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CHEMISTRY AND MICROBIOLOGY OF GREEN BUILDING MATERIALS

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CHEMISTRY AND MICROBIOLOGY OF GREEN BUILDING MATERIALS

by

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In dedication to my Mum who always wants the best for her children

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CHEMISTRY AND MICROBIOLOGY OF GREEN BUILDING MATERIALS

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While the market for "green" building materials has been expanding rapidly, no rigorous framework exists for evaluating the chemical and biological reactivity of these building materials. The objective of this research was to assess the ozone reactivity, primary and secondary VOC emission rates and mold resistance of selected green building materials. Two different sets of experiments were conducted. The first set focused on reactive consumption of ozone by ten common green materials. A screening assessment of secondary emissions of C_6 and greater carbonyls was also completed for selected green materials. The second set was completed to evaluate the relative resistance of selected green building materials and their conventional analogs to surface fungal growth in moist interior environments.

Ozone reactivity varied considerably between test materials. The ozone deposition velocity for inorganic ceiling tiles, for example, was two times higher than cabinetry materials and approximately fifty times higher than UV-coated bamboo. Experimental results were used as input to a simple mass balance model which predicted that the ratio of indoor to outdoor ozone concentrations was not significantly affected by green building materials. The green materials used in this study emitted less primary and secondary VOCs than did their non-green counterparts, although the difference was not significant and the material sample set was relatively small. Also, the green materials tested were not prone to either less or more mold growth than their conventional counterparts. Instead, materials composed of organic materials with high equilibrium moisture contents (EMC) were more prone to mold growth than inorganic materials with low EMC. Perlite-based (inorganic) ceiling tiles that consumed relatively large amounts of ozone without corresponding by-product formation were also resistant to mold growth. Such findings should facilitate the selection of future green building materials, both explicitly and by defining a protocol for future testing of green materials.

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1. INTRODUCTION

1.1. Problem Statement

Total human exposure to air pollution is dominated by the amount of time that most people spend indoors and the relatively high levels of air pollution present indoors [1]. Buildings can have a significant impact on occupant health, and indoor building materials can be a major source of volatile organic compounds (VOCs), bioaerosols, and particulate matter in interior environments [2-7]. Composite wood materials, for instance, are common in indoor environments and can produce formaldehyde, and biological pollutants such as fungal spores [2, 3, 7]. Recently, there has been a rapid increase in the marketing and utilization of green building materials in response to rising energy costs and concerns over global warming and indoor air quality. While there has been extensive research to delineate the chemical and biological reactivity of conventional building materials, the reactivity of green building materials has been virtually unstudied. Indeed, many materials are touted as being "green" without a robust scientific definition of the term. Thus, there is a need to better define the criteria for green building materials and to delineate the reactivity of such materials in the indoor environment.

In order to facilitate the selection of future green building materials, the physical, chemical and biological properties of these materials must be considered (Figure 1). These properties all relate to each other in one way or another. For instance, hydrophilic materials may be more prone to mold growth, or mold infested materials may become more reactive with ozone.

One criterion, sometimes used for green materials, is that they are "low VOC-emitting" [8, 9]. However, this criterion generally applies to primary emissions, typically of VOCs, which are emitted from the actual components of the manufactured product [10]. These green materials may contain components that undergo significant reactions with ozone. High ozone reactivity might be considered a positive from the standpoint of removing ozone from indoor air, but a negative if such reactions have an adverse effect on the materials or if they produce irritating or toxic by-products such as carbonyls. Thus, measuring primary emissions of indoor materials alone may not be sufficient, since secondary emissions that are generated from ozone reactions with building materials may dominate over a product's life-time [10]. In addition, many green materials are bio-based,

and made of components that may be more conducive to fungal growth than conventional building materials.

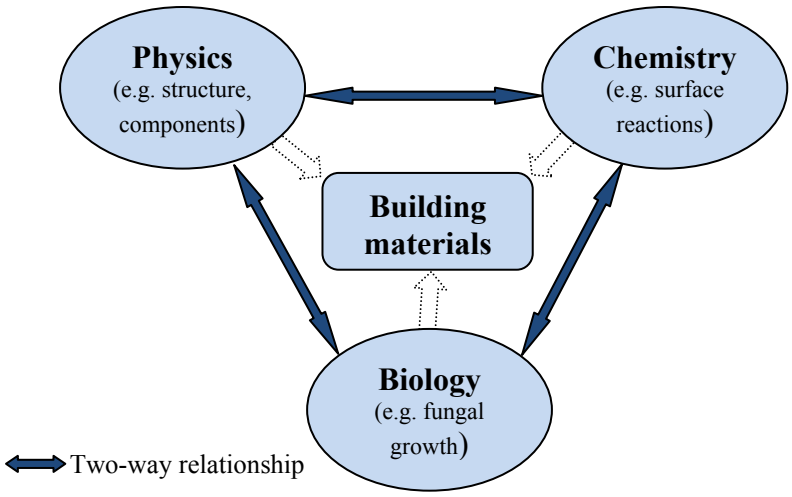


Figure 1. Physical, chemical, & biological characteristics of materials

This study involved an analysis of the biological and chemical characteristics of building materials in terms of their ozone reactivity and fungal resistance. Past research has focused on conventional building materials, while there is little published literature related to the chemical and biological reactivity of green materials. It would be valuable to identify green materials that have the following positive attributes: (1) high removal of ozone from indoor air, (2) small or no formation of reaction products, and (3) high resistance to mold growth.

1.2. Objectives

This dissertation focused on (1) ozone removal by green building materials, (2) comparison of secondary emissions generated from ozone reactions with selected green building materials and non-green counterparts, and (3) the ability of these materials to sustain fungal growth. More specifically, the objectives of this study were to:

1. Quantify the deposition velocity and reaction probability of ozone for a range of green building materials.
2. Identify by-products resulting from reactions between ozone and building materials.
3. Quantify the release rate of selected VOCs from each green building material following exposure to ozone.

4. Evaluate the effects of nutrients, spore levels, and surfactants on the growth of a model fungus on selected green materials and their conventional analog materials.
5. Compare the resistance of selected green materials and their conventional counterparts to colonization by common indoor fungi following water saturation or exposure to high humidity conditions.

1.3. Scope

The study was conducted in three major phases as follows:

- ***Phase I: Ozone removal.*** The deposition rates and reaction probabilities of ozone with ten selected green materials at a relative humidity of 39 -56%, temperature of 22-24^{o}C, and an air exchange rate of 1 h^{-1} were determined. Experiments were completed in small (48-L) flow-through laboratory chambers.
- ***Phase II: By-product formation.*** A set of four different green materials and their non-green counterparts were selected to quantify emissions of saturated aldehydes (C_6-C_{10}) from ozone reactions with test materials. Lighter carbonyls and other oxidized products, such as carboxylic acids, were not measured due to instrument limitations during this study. Experiments were completed in small (48-L) flow-through laboratory chambers.
- ***Phase III: Fungal resistance.*** Four pairs of green/non-green materials were selected for fungal growth experiments. Two different methodologies were employed to inoculate the materials: (1) materials were artificially inoculated with serial dilutions of *Aspergillus niger (A. niger)* spores suspended in a range of nutrient solutions, and (2) materials were naturally inoculated by exposing them to an indoor environment containing a mixture of fungi. Mold growth was evaluated periodically over two months of incubation at a constant temperature of 30^{o}C and RH of 90-95%. Experiments were completed in Sanpla dry keeper cabinets (Sanplatec Corp., Japan).

1.4. Organization

This dissertation is organized into an executive summary followed by the complete text (Appendices 1-3) for three supporting papers that have either been accepted

for publication or that will soon be submitted to journals for review (see section 1.5). The executive summary has been divided into six descriptive sections as follows:

- Section One introduces the problem statement and research objectives, which focus on evaluating the chemical and biological characteristics of green materials with respect to ozone reactivity and fungal resistance.
- Section Two provides background information on the existing green building rating program and identifies unanswered biological and chemical concerns that may affect the selection of future building materials.
- Section Three summarizes the experimental methodology utilized to evaluate ozone removal and by-product formation from ozone reactions with selected green materials and/or their non-green counterparts. Key findings are provided at the end of the section.
- Section Four describes the methodology employed to assess the fungal resistance of green and conventional building materials and discusses key findings.
- Section Five provides a cross-comparison of the biological and chemical reactivity of four selected pairs of green and non-green materials with respect to three criteria: ozone removal, by-product formation, and fungal resistance.
- Section Six summarizes the key conclusions for the research and identifies future research needs.

1.5. List of Journal Papers

This dissertation is based on the following papers, which will be referred to as Appendices 1 to 3. These papers are attached at the end of this dissertation.

1. Chi P. Hoang, Kerry A. Kinney, and Richard L. Corsi. Ozone removal of green building materials, *Buildings and Environment (2009)*. Vol 44, Pages: 1627-1633.
2. Chi P. Hoang, Kerry A. Kinney, Richard L. Corsi, and Michael Waring. Secondary emissions from ozone reaction with green building materials, (to be submitted as a technical note).
3. Chi P. Hoang, Kerry A. Kinney, Richard L. Corsi, and Paul J. Szaniszlo. Resistance of green building materials to mold growth (to be submitted soon).

2. GREEN BUILDING MATERIALS

Buildings have a significant impact on occupant health, the economy, the environment, and even on the surrounding ecosystem, since many building activities from design and construction through maintenance involve significant consumption of energy and natural resources (40% of total U.S. energy consumption) [11]. As the cost of energy rises and concerns over green house gases and global warming increase, there is an increasing interest in green buildings. The term "green" is used increasingly to mean environmentally friendly [11, 12]. More specifically, Spiegel and Meadows (1999) define a green building as a building that is located and constructed to allow its occupants to live, work and play in a sustainable manner [12]. The U.S. Environmental Protection Agency has listed some potential benefits of green buildings [11]. These include various environmental benefits:

- Enhance and protect biodiversity and ecosystems.
- Improve air and water quality.
- Reduce waste streams.
- Conserve and restore natural resources.

Over the past fifteen years, interest in green issues in the United States has grown considerably. In 1991, there was only one local program in the country giving market recognition to green builders and their homes - the city of Austin's Green Builder Program. In 1998, at least seven new programs were established, including Build A Better Kitsap Home Builder Program and the County of Santa Barbara Innovative Building Review Program [13]. As of 2002, more than 18,000 green homes have been constructed and certified as green homes. In 2005, there were 28 green home building programs in operation throughout the United States [14]. These increasing numbers imply an existing and developing new market demand for more resource-efficient homes. Along with the increasing interest in homes that are better for the environment and occupant health, the U.S. Green Building Council (USGBC) introduced the Leadership in Energy and Environmental Design (LEED) Program in 1993 in order to develop a new generation of buildings that utilize energy, water and natural resources in an efficient manner and provide a healthy built environment for occupants [15].

Green building materials are one of the most important components of green buildings. These materials are intended to be environmentally friendly with such characteristics as low toxicity, minimal chemical emissions, high recyclability, and long durability [12]. In the LEED green building rating system, materials are among six important categories for sustainable design [15]. To help customers and builders select green products, several directories of green building products and materials are available [9, 12, 16]. However, the issue of which building materials are better from an environmental standpoint is still controversial. In compiling any directory of green building products, the authors have to determine what qualifies a product for inclusion. It is important to note that many of the criteria used to select green products are subjective, and a product may perform well under one criterion, but poorly under another. Tradeoffs between different criteria are inevitable. Therefore, once a potential product is identified, that product is carefully assessed for its "greenness." The process used to select products for inclusion in the GreenSpec directory, a comprehensive list of green building products, is based on six main criteria [9]:

- products that are made with salvaged, recycled, or agricultural waste content.
- products that conserve natural resources.
- products that avoid toxic and other emissions.
- products that reduce environmental impacts during construction, demolition, or renovation.
- products that save energy and water.
- products that contribute to a safe, healthy indoor environment.

Many green materials are bio-based and may consist of unsaturated acids that are highly reactive with ozone (and perhaps also a significant "secondary" emission source of reaction products such as aldehydes and ketones). These green materials also may be more prone to mold growth. Therefore, studying the ozone reactivity of green building materials and assessing their susceptibility to fungal growth are important first steps toward better assessing the potential impact of these materials on the quality of the indoor environment.

3. OZONE REACTIONS WITH GREEN BUILDING MATERIALS

3.1. Background

Heterogeneous (surface) reactions between ozone and conventional building materials have been studied by several research teams [17-22]. Heterogeneous reactions are typically quantified in terms of a deposition velocity or, more fundamentally, a reaction probability. Ozone deposition velocity onto materials, defined as the flux of ozone to a surface divided by its mean concentration in air, depends on two process resistances: transport resistance ($r_{t,}$ s/m), and surface uptake resistance ($r_{s,}$ s/m) [22]. The reaction probability is defined as the number of reactions that occur per number of collisions of a reactive molecule, e.g., ozone, with a material. For small reaction probabilities, the surface uptake process is rate-limiting, while transport processes can be rate limiting in the case of high reaction probabilities. Several authors have provided insightful discussions of the meanings of deposition velocity and reaction probability, and the relationship between the two [18-24]. An additional discussion of deposition velocity and reaction probabilities is provided in Appendix 1.

In addition to heterogeneous reactions, indoor ozone concentrations are dependent on four other factors: (a) outdoor ozone concentration, (b) building or zonal air exchange rate, (c) indoor emission sources of ozone, and (d) homogeneous reactions in room air. Assuming well-mixed conditions with first-order homogeneous and heterogeneous reactions, a mass balance for ozone can be expressed as:

$$V\frac{dC}{dt} = QC_o - QC + E - \sum v_{d,i}A_iC - \sum_R k_R V C_R C \qquad (1)$$

where Q is the volumetric air flow rate through a building (m^3/hr); C and C_o are the indoor and outdoor concentrations of ozone, respectively (μg/m^3); E is the indoor emission rate of ozone (μg/hr), V is the interior volume of a well-mixed building or zone with a building (m^3), k_R is the bimolecular reaction rate constant for compound R and ozone (m^3/μg/hr), C_R is the indoor concentration of reactant compound R (μg/m^3); $v_{d,i}$ is the deposition velocity for ozone to materials *i* (m/hr); and A_i is the interior surface area for material *i* (m^2).

In the absence of indoor sources (E ≈ 0) and if heterogeneous reactions are of much greater significance than homogeneous reactions with respect to ozone removal,

Equation 1 can be solved at steady-state for the ratio of indoor-to-outdoor (I/O) ozone concentrations:

$$I/O = \frac{C}{C_0} = \frac{1}{1+\frac{\sum_{i=1}^{n} v_{d,i} A_i}{\lambda V}} = \frac{1}{1+\frac{v_d A}{\lambda V}} \quad (2)$$

As shown in Equation 2, the summation of heterogeneous reactions over all *n* materials in a building are often simplified and represented by single area-averaged deposition velocity (v_d) acting over the entire indoor surface area (A).

Building materials are usually the largest contributor to total indoor surface area, and are one of the major contributors to indoor VOCs. This is particularly true for new materials. For instance, Zhang et al. [2] stated that 60% of total volatile organic compounds (TVOCs) originate from building materials and furnishings. Wolkoff (1999) grouped emissions from building materials into two categories: primary and secondary emissions [10]. Primary emissions refer to the release of free and non-bound VOCs that are originally in gas or physically adsorbed phases within porous materials [10]. Secondary emissions, resulting from chemically and physically bound VOCs, are produced from interactions of materials with external parameters, such as heat, light, and strong reactive chemicals [9, 25-27]. Nui and Burnett (2001) listed chemicals that are emitted from some common building materials [28]. For example, formaldehyde and terpenes were emitted primarily from particle board and wood products while carpet and paint were considered major sources for various VOCs [2, 19, 28]. James and Yang (2005) conducted a study to compare primary emissions of three pairs of green and non-green materials, including Trex® versus pressure treated wood, ceramic floor tile versus vinyl composite floor tile, and water-based versus oil-based paints [8]. They found that green materials emitted lower quantities of VOCs than did their non-green counterparts.

A few other researchers have measured the formation of secondary emissions from ozone reactions with building materials. Hyttinen et al. (2007) studied the oxidation products of clean and used HVAC filters with ozone [29]. They observed no change in ozone concentration when ozone passed through a clean fiberglass filter. On the contrary, the dust collected on the filters during their service period had components that reacted with ozone. Consumption of ozone was observed in almost all used filters. Morrison and

Nazaroff (2002) measured secondary emissions from carpets and found an increase in some aldehydes, such as nonanal after carpets were exposed to ozone [19]. Poppendieck et al. [18, 30] studied by-product formation from 24 materials during and after a disinfection period in which the materials were exposed to 1,000 ppm of ozone. The largest total by-product mass releases were observed for three materials - paper, office partitions, and medium density fiberboard. They also found that by-product formation after material ozonation can persist for months or even longer [18, 30].

3.2. Overview of Experimental Procedure

This portion of the research was intended to address Objectives 1-3 (see Section 1.2). Two different sets of experiments were conducted to evaluate ozone reactivity and by-product formation from selected building materials.

3.2.1. Ozone Reactivity

Ten green building materials were selected for this study, including inorganic (perlite-based) ceiling tile, unglazed ceramic tile, natural cork wall-paper, aluminum tinted cork wall-paper, bamboo, UV-coated bamboo, wheat board, UV-coated wheat board, sunflower board, and UV-coated sunflower board. All materials were unused when obtained; either shipped directly from manufacturers (ceiling tiles, ceramic tiles, and cork wall-coverings) or donated by a green builder in Austin, Texas (bamboo, wheat board, and sunflower board). Upon collection, materials were wrapped in multiple layers of plastic sheeting and stored for periods of up to several weeks before an experiment. Test materials and abbreviations used in this study are listed in Table 1.

The experimental system consisted of two 48-L electro-polished stainless steel chambers operated in parallel. Prior to each experiment, the chambers were cleaned with deionized water and dried with a heat gun in order to remove gases that had adsorbed onto, or particles deposited on, the chamber walls. Ozone was generated by use of UV light. The relative humidity of the supply air was adjusted by use of a split stream; one stream was passed through a water column (humidifier) before being reconnected to the main stream. Ozone concentrations in the inlet and exhaust streams of each chamber were determined by using single UV-cell ozone monitors (2B Technologies, model 202).

The mean inlet ozone concentration varied between experiments (100 ppb to 150 ppb). For the standard (base case) condition, temperature was maintained at between 22°C and 24°C, while relative humidity was controlled at 39% - 56%. The effects of a higher relative humidity of 70% were also studied for four green materials (natural/aluminum cork wall-coverings, ceiling tiles, and ceramic floor tiles). During each experiment, chambers were continuously ventilated at an air exchange rate of 1 hr^{-1}. Each experiment was divided into three separate experimental phases. The first phase began when ozone was injected into the experimental system for a period of 48 hours. In the second phase, the UV lamp was switched off to discontinue the ozone supply to the chamber for a period of one day. After one day, the UV lamp was switched back on, and ozone was again injected into the chambers to assess whether the reactivity of the material had been regenerated.

Table 1. Test materials, designations, and applications

Material	Designation	Manufacturer/Model	Indoor application
Unglazed ceramic tiles	Ceramic	Fireclay Tile/Debris Series	Flooring
Perlite-based ceiling tiles	Ceiling	Chicago Metallic/Novum	Ceiling
Unfinished bamboo	Bamboo	Smith & Fong/Plyboo	Flooring and/or others
UV-coated bamboo	Bamboo_UV	Smith & Fong/Plyboo	Flooring and/or others
Unfinished sunflower	Sunflower	Environ Biocomposites/Dakota Burl	Cabinetry and/or furniture
UV-coated sunflower	Sunflower_UV	Environ Biocomposites/Dakota Burl	Cabinetry and/or furniture
Unfinished wheat	Wheat	Environ Biocomposites/Biofiber™ Wheat	Cabinetry and/or furniture
UV-coated wheat	Wheat_UV	Environ Biocomposites/Biofiber™ Wheat	Cabinetry and/or furniture
Natural cork	Cork_Na	Innovations, in Wall-coverings/Invironmentals	Wall covering
Aluminum tinted cork	Cork_Alum	Innovations, in Wall-coverings/Invironmentals	Wall covering

To determine reaction probability, material samples were coated with a concentrated solution of sodium nitrite [1g of sodium nitrite ($NaNO_2$), 1g of potassium carbonate, 2 ml of glycerol in a solvent of 70 ml deionized water and 30 ml of methanol] and allowed to dry at room temperature. The same experimental procedure as described above was applied to $NaNO_2$-coated materials.

The time varying deposition velocity for each material was estimated by solving Equation 1 for v_d with analysis of consecutive pairs of ozone concentrations as shown in Equation 3:

$$v_d(t=t_{ave}) = \frac{2/\Delta t\left[C^n - C^{n-1}\right] + \lambda\left[C_0^n + C_o^{n+1} - \left(C^n + C^{n+1}\right)\right]}{A/V\left[C_o^n + C_o^{n+1}\right]} - v_{d,c}\left(t=t_{ave}\right)\frac{A_c}{A} \quad (3)$$

Here, the superscript "n" corresponds to successive time steps, and all other variables are as described previously. The deposition velocity was taken at a time calculated as the mid-point (linear average, t_{ave}) between consecutive ozone measurements. The time increment (Δt) corresponded to the time between successive ozone measurements. To determine the ozone deposition velocity associated with the chamber walls ($v_{d,c}$), the same experimental procedure described in the previous section was carried out for both empty chambers. Additional details related to reactivity experiments or uncertainty calculations are presented in Appendix 1.

3.2.2. By-Product Formation

The concentration of by-products from ozone reactions with four different pairs of green and non-green building materials (bamboo vs hardwood flooring, sunflower board vs particle board, inorganic vs non-green ceiling tiles, paperless drywall vs non-green gypsum board) exposed to ozone was assessed using the same experimental system as described above for reactivity experiments. Each experiment consisted of three-phases. Phase 1 (pre-exposure) involved the placement of a material in a test chamber for 8 h prior to exposure to ozone. Gas-phase samples were taken to quantify the release of heavy carbonyls (C_6 and greater saturated carbonyls) associated with primary emissions from each material. During Phase 2 (ozone exposure), ozone was introduced to each chamber for 6 hours. By-product measurements were not made during this phase. During Phase 3 (post exposure) ozone was no longer introduced in the inlet air. By-product samples were collected 4, 24, and 72 h after the termination of ozone injection in the chamber inlet. Sample air from the chambers was drawn through Tenax-TA ™ adsorbent tubes and analyzed by GC/MS. Details related to the sampling and analysis procedure and related uncertainty estimation are provided in Appendix 2.

3.3. Key Findings

3.3.1. Ozone Removal

The time-dependent ozone deposition velocities (with error bars) for nine different green materials and electro-polished stainless steel are presented in Figure 2. As expected, the deposition velocity for ozone onto electro-polished stainless steel reached a constant and relatively small value after the first five hours. Deposition velocities measured for ceiling tile, natural and aluminum-tinted cork wall boards, wheat board and sunflower board were much greater than the deposition velocities determined for the other green materials tested in this study.

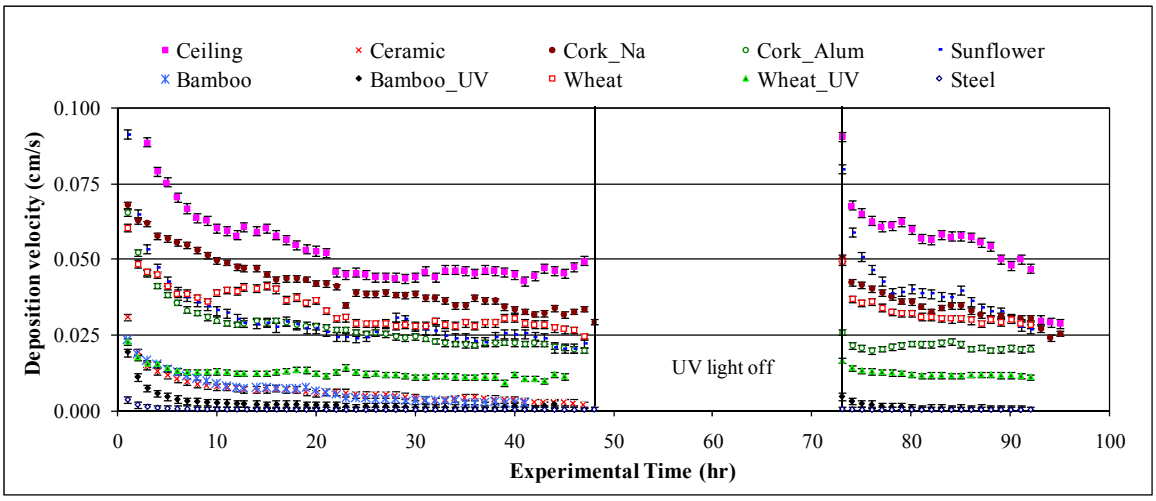


Figure 2. Time dependent deposition velocities for green building materials

For each material, the deposition velocity decreased rapidly during the first 10 h, before it leveled off to an approximately constant value. During the second exposure phase, a regeneration of ozone deposition velocity was observed, followed by a second decay in deposition velocity. Regeneration was particularly pronounced for ceiling tile and sunflower board. A rapid decay in deposition velocities was likely due to a consumption of sites that are highly reactive with ozone. These sites might be associated with the materials itself or with particles that deposit on, or gases that adsorb to, materials. The regeneration of reaction sites on test materials after 24 hr without ozone exposure is important. Such regeneration was likely not due to deposition of particles or adsorption of gases onto the materials during the 24-hr-period without ozone exposure,

since test materials were contained in experimental chambers with a conditioned inlet air stream as described above. Instead, it is conceivable that reaction sites on material surfaces were consumed during ozone exposure, thus establishing a concentration gradient between reactive molecules in the test material matrix and the surface of the material. This would induce molecular diffusion of such molecules to the surface, with effective surface replenishment (or regeneration) of reaction sites. During experiments the deposition velocity decays but levels off as the rate of diffusion to the surface approaches the rate of consumption of reaction sites by ozone at material surfaces. This is a potentially important phenomenon given variations in outdoor ozone (a major source of indoor ozone) and intermittent usage of indoor sources of ozone, e.g., daytime ion generators, laser printers, and photocopy machines.

For the purpose of assessing ozone deposition velocities among different materials, and also for comparison to previous research, time-dependent deposition velocities were averaged for the last day of the first exposure phase when deposition onto test materials had approximately leveled off. The ozone deposition velocities between test materials differed by approximately a factor of 50, from 0.001cm/s for UV-coated bamboo to 0.046 cm/s for ceiling tile.

Importantly, the deposition velocities of the same material, with and without a coating or pigment, were considerably different for wheat board, cork, and bamboo. Materials with coatings or pigments yielded lower ozone deposition velocities, presumably due to coverage (shielding) of reaction sites by a solid coating or pigment particles that are less reactive with ozone. Two cork wall-covering products, for example, had ozone deposition velocities of 0.026 cm/s (aluminum-tinted cork) and 0.037 cm/s (natural cork). This is similar to the trend observed for wheat board and UV-coated wheat board, and for bamboo and UV-coated bamboo.

The ratios of transport limited resistance to total (transport and surface reaction) resistance are presented in Table 2. Ceramic tile, bamboo, UV-coated bamboo, aluminum tinted cork, and UV-coated wheat yield a resistance ratio of less than 0.1; surface reaction processes are dominant in terms of ozone deposition onto these materials. In contrast, the resistance ratios for ceiling tile, natural cork, wheat and sunflower board range from 0.3 to 0.5, indicating that both transport processes and surface reactions play key roles in

ozone removal to these test materials. Ceiling tile had the largest reaction probability (≈ 10^{-5}), an order of magnitude or more greater than the reaction probabilities for bamboo, UV-coated bamboo, and ceramic materials (Figure 3).

Table 2. Deposition velocities and relative resistances for ozone removal to test materials

Material	Deposition velocity (cm/s)		Resistance (s/cm)		Resistance ratio $r_t/(r_s+r_t)$
Ceiling	v_d	0.046	r	22	0.500
	v_t	0.091	r_t	11	
	v_s	0.091	r_s	11	
Ceramic	v_d	0.004	r	255	0.054
	v_t	0.072	r_t	14	
	v_s	0.004	r_s	241	
Bamboo	v_d	0.003	r	296	0.051
	v_t	0.066	r	15	
	v_s	0.004	r_t	281	
Bamboo_UV	v_d	0.001	r_s	875	0.022
	v_t	0.053	r	19	
	v_s	0.001	r_t	856	
Sunflower	v_d	0.025	r_t	40	0.277
	v_t	0.090	r_s	11	
	v_s	0.034	r	29	
Sunflower_UV	v_d	0.021	r_t	47	0.190
	v_t	0.112	r_s	9	
	v_s	0.026	r	38	
Cork_Na	v_d	0.036	r_t	28	0.303
	v_t	0.118	r_s	8	
	v_s	0.051	r	20	
Cork_Alum	v_d	0.023	r_t	44	0.088
	v_t	0.259	r_s	4	
	v_s	0.025	r	40	
Wheat	v_d	0.028	r_t	35	0.391
	v_t	0.072	r_s	14	
	v_s	0.047	r	21	
Wheat_UV	v_d	0.011	r_s	89	0.094
	v_t	0.119	r	8	
	v_s	0.012	r	81	

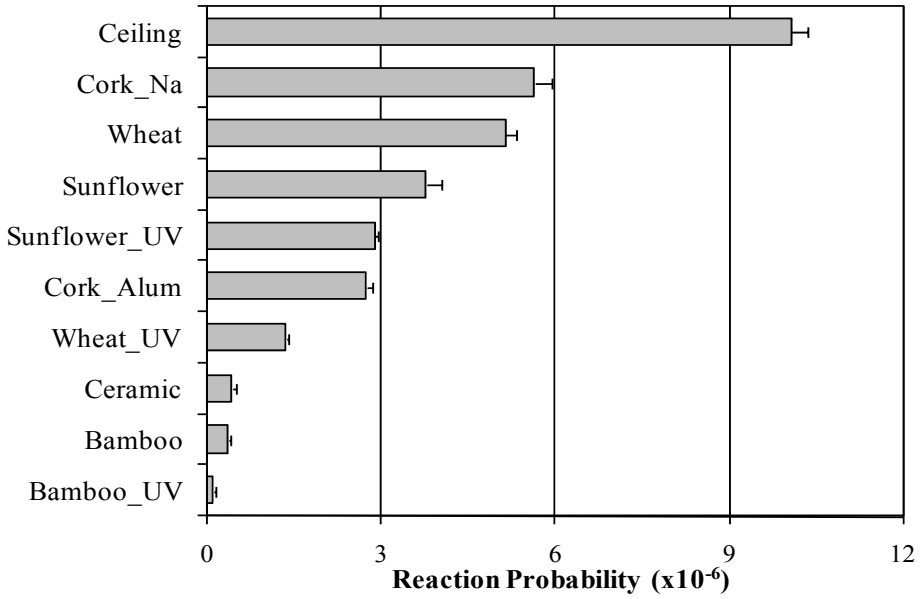


Figure 3. Reaction probability for green building materials

To evaluate the effects of several green building materials on the I/O ratio for ozone, a simple well-mixed reactor model was employed under assumed steady-state conditions. The model was applied to an empty test room (no furniture, etc.) with dimensions of 10m x 10m x 3m (length x width x height). Painted gypsum board, carpet, and common ceiling tiles were used as conventional indoor materials for the base case test room. Ozone deposition velocities for latex painted gypsum board and ceiling tiles are available in the literature [18, 31]. For this assessment, a value of 0.042 cm/s was used for painted gypsum board [31] and 0.07 cm/s was selected for ceiling tile [18]. Numerous deposition velocities have been reported for carpet, with a significant range of values [31]. For this study, a range of 0.032 cm/s to 0.7 cm/s was selected for carpet [31]. Deposition velocities observed in this study were employed for perlite-based ceiling tiles, aluminum tinted cork wall coverings, and UV-coated bamboo flooring to explore their effects on I/O ratios. Specific modeling scenarios are listed in Table 3.

The results of indoor-to-outdoor (I/O) ratios of ozone concentration for the various scenarios are presented in Figure 4. Air exchange rate (λ in Equation 2) has a significant effect on I/O ratios, with the indoor ozone concentration decreasing with decreasing λ. In the presence of carpet, increasing λ from 0.2 hr^{-1} to 2 hr^{-1} doubles the I/O

ratio. The I/O ratio also depends on the surface deposition of ozone onto different materials. For example, compared to the base case (with low reactive carpet), altering conventional materials with their green counterparts does not affect the I/O ratio much. However, if highly reactive carpet is used for the base case, the I/O ratio for scenarios involving UV-coated bamboo flooring increases by 50% with low λ to 10% with high λ.

Table 3. Scenarios used to estimate indoor/outdoor ozone concentrations for test room

Scenario	λ	A/V			Materials		
		Ceiling	*Wall*	*Flooring*	*Ceiling*	*Wall*	*Flooring*
1	0.2 - 2	1/3	2/5	1/3	Ceiling tile	Painted gypsum	Carpet
2	0.2 - 2	1/3	2/5	1/3	Green Ceiling tile	Painted gypsum	Carpet
3	0.2 - 2	1/3	2/5	1/3	Ceiling tile	Aluminum tinted	Carpet
4	0.2 - 2	1/3	2/5	1/3	Ceiling tile	Painted gypsum	UV-coated bamboo
5	0.2 - 2	1/3	2/5	1/3	Green Ceiling tile	Aluminum tinted	UV-coated bamboo

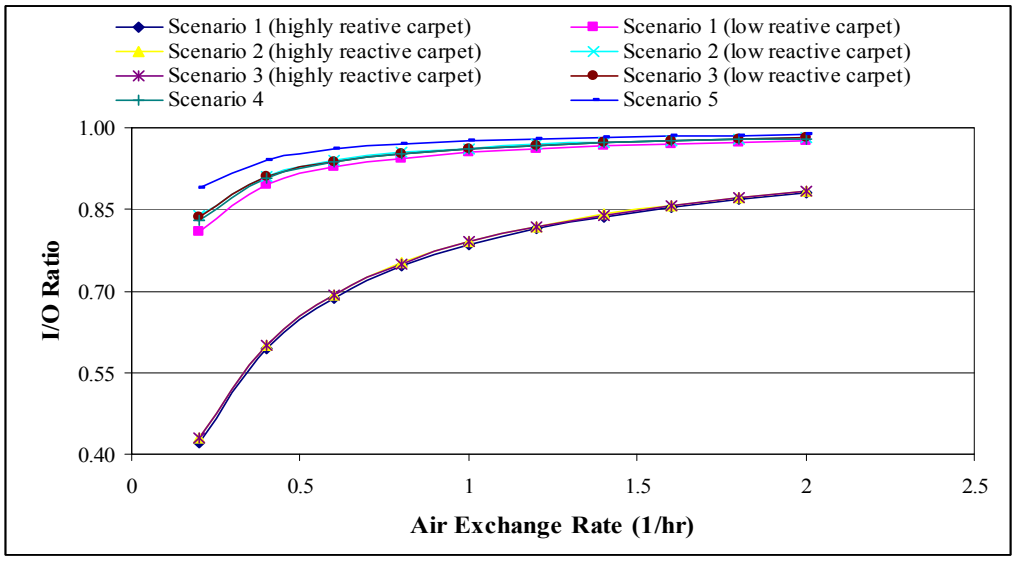
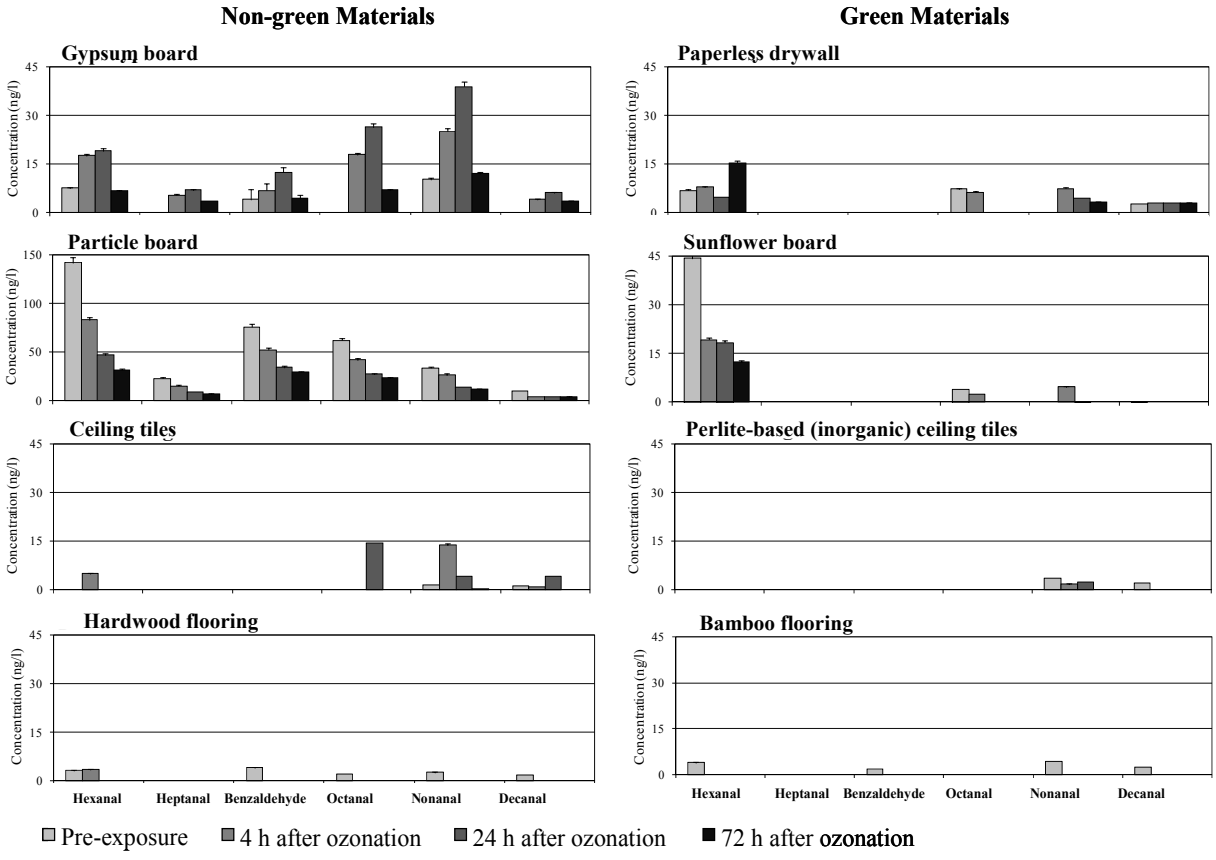


Figure 4. I/O ratio for different material configurations

3.3.2. VOC Emissions

Chamber headspace concentrations associated with primary and secondary VOC emissions from four selected green materials and their non-green counterparts (bamboo vs hardwood flooring, sunflower board vs particle board, inorganic (or perlite-based) vs non-green ceiling tiles, paperless drywall vs non-green gypsum board) were studied. Among 12 target compounds selected for analysis, only six aldehydes, including hexanal, heptanal, benzaldehyde, octanal, nonanal, and decanal, were detected. Results are summarized in Figure 5.



* The concentration scale for particle boards is 0-150 ng/l while others are from 0-45 ng/l

Figure 5. By-product formation from green and non-green building materials

Particle board produced the highest carbonyl concentrations in chamber air. Sunflower board was characterized by a total carbonyl concentration an order of magnitude lower than that of particle board. However, among four selected green materials, sunflower board was the largest contributor of primary and secondary

carbonyls. This observation is likely due to the fact that sunflower board contains unsaturated fatty acids, such as oleic and linoleic acids, which can react with ozone leading to the formation of several oxygenated compounds such as hydroperoxides, ozonides, and aldehydes [27, 32]. Interestingly, the concentrations of all identified compounds, observed from these two cabinetry materials, declined significantly just four hours after ozonation and continued declining slowly until the end of the sampling period. For example, the concentration of hexanal from sunflower board dropped from 45 μg/L (before ozonation) to slightly lower than 20 μg/l (4 hours after being exposed to ozone) and slowly decreased to about 13 μg/L at the end of the sampling period. This observation was also reported in several other studies of conventional materials [18, 30]. One possible reason for these observations is that ozone may react with compounds that were not quantified in this study and formed hydroxyl radicals (OH*), which then rapidly reacted with carbonyls associated with primary emissions to produce carboxylic acids and other by-products that were not quantified in this study. Pine-based particle board, for example, emits significant amounts of α-pinene, which is known to react rapidly with ozone to form hydroxyl radicals [33].

Wall-board materials, gypsum board, and drywall, emitted similar amounts of identified carbonyls prior to ozone exposure. However, gypsum board clearly emitted greater quantities of carbonyls following exposure to ozone, although the paperless drywall did emit slightly more hexanal and nonanal after ozone exposure than before. It is not clear as to whether the increased emissions from gypsum board stemmed from reactions with the paper surface or underlying gypsum.

Both inorganic ceiling tiles and bamboo flooring were characterized by low carbonyl concentrations before and after ozonation. Almost no chemicals were emitted from bamboo flooring even after the ozonation period. Inorganic ceiling tile exhibited a large ozone deposition velocity but produced little carbonyls, even after six hours of ozonation. This finding underscores the possibility of identifying materials that remove substantial amounts of ozone from air (a positive attribute) while forming little to no reaction products (another positive attribute). Additional information related to by-product formation is provided in Appendix 2.

4. MOLD GROWTH ON GREEN BUILDING MATERIALS

4.1. Background

In recent years, mold growth in buildings has attracted public interest, as the billions of dollars spent on mold related repairs and litigation [34], and adverse effects of fungal bio-aerosols on building occupants [7, 35-36] have been well documented. Approximately 40% of study homes in six U.S. cities have been affected by mold [37]. In California alone, an estimated $364 million has been spent to repair mold damage in homes [38]. In addition, there is currently a substantial body of evidence to support the viewpoint that fungi in buildings can have severe and wide ranging effects on the general health of occupants [39-41]. Mold spores, for example, can become airborne and can produce health problems such as asthma and allergies. Nielsen et al. (2004) classified health problems associated with moldy and damp buildings into three categories: mucosal symptoms, lung symptoms, other general symptoms [36]. The health effects observed in moldy buildings may be linked to the fact that fungi can produce microbial fragments and volatile organic compounds which may cause allergies or asthma and generate unpleasant odors [42-50].

Mold and other fungi can be found in many places, e.g., in soil, food, and building materials. Major factors that regulate microbial growth include nutrients, moisture, and temperature. While the use of biocidal compounds may be appropriate to limit mold growth in new buildings and to alleviate existing problems, the preferred and most effective strategy is to eliminate the conditions that can lead to fungal growth [41]. By understanding parameters affecting mold growth within building materials, it may be possible to prevent mold growth effectively.

4.2. Overview of Experimental Procedure

Although several studies have examined fungal growth on conventional materials in dry and wet environments, there is little published work related to the biological reactivity of green materials. For this reason, the objective of this study was to compare the susceptibility of green building materials and their non-green counterparts to fungal colonization in simulated interior environments. More specifically, the question of how fast fungi grow on interior material surfaces under nearly-water-saturated conditions

and/or following high humidity exposure (e.g., such as would occur when building structures are saturated with water during a flooding event) needs to be answered for green materials. In order to provide guidance on the selection of building materials used for moisture sensitive locations, this research focuses on evaluating the relative resistance of green building materials and their conventional analogs to surface fungal growth in severe interior environments.

Briefly, two major sets of experiments were conducted using four pairs of green and non-green building materials. In the first set of experiments, an artificial inoculation protocol was used to investigate the effects of external nutrient levels, host materials, spore levels, and surfactants on the susceptibility of test materials to a model fungal species. The second set focused on a natural inoculation protocol that was conducted to evaluate the resistance of a limited number of green and non-green building materials to colonization by common indoor fungi after direct water exposure and/or following high humidity exposure. Detailed experimental procedures are provided in Appendix 3.

4.2.1. Artificial Inoculation

The purpose of this task was to assess the effect of spore concentration, external nutrients on the growth rate of a model fungus on the surfaces of both green materials and their non-green counterparts. *Aspergillus niger* (*A. niger*) was selected as the model species for the study. The surface of each building material to be tested was inoculated with *A. niger* spores mixed with one of the following four nutrient solutions: sterile de-ionized water (containing no nutritive components and no vitamins for the cultivation of yeast and molds); yeast nitrogen base (containing all nutritive components and vitamins, except for a carbon source); yeast carbon base (containing all nutritive components and vitamins, except for a nitrogen source); yeast base (containing all nutritive components and vitamins). Each spore solution (10 μl) was diluted to achieve from 10 to 10^5 spores/inoculated spot on the surface of each pre-sterilized building material sample (See Appendix 3 for more details). After inoculation, the building material samples (8 cm x 12 cm) were incubated for a period of two months in a conditioned chamber at a constant temperature of 30^{o}C and RH of 90-95% (controlled by a saturated K_2SO_4 solution).

In order to evaluate the effects of different levels of nutrients on the growth of *A. niger*, 1X and 10X strength nutrient solutions were used to dilute the spores (Experiment 1 and 2, respectively). Spore-free solutions were also applied to the material's surfaces to serve as negative controls to indicate whether unintentional microbial contamination of the specimens occurred during the experiments., The time until growth was first observed with a stereo microscope (Olympus SZ-ST, Olympus America Inc., Center Valley, PA, U.S.A) was used to compare the resistance of the test materials to mold growth for each growth solution, and spore concentration. The longer the elapsed time between the first observation of growth and the time of inoculation, the more resistant the material was considered to be to mold growth.

4.2.2. Natural Inoculation

The purpose of this test was to investigate the growth rate of various common fungi on the surfaces of both green materials and their non-green counterparts. After sterilization, the test materials were left unprotected in a residential house to allow natural fungal inoculation over a period of 10 days. The material samples were not inoculated with additional specific fungal species; thus, this method is termed natural inoculation. Two different sets of experiments were carried out:

1. **Direct water exposure**: material specimens were first submerged in sterile water for ten hours to simulate a direct exposure to water; then incubated in a conditioning chamber at a constant temperature of 30^{o}C and RH of 90-95%.
2. **High humidity exposure**: the experimental procedure used was the same as that described above, with the exception that the materials were not submerged in water before incubation.

Negative controls for both sets of experiments were material samples submerged in sterile water for ten hours but not exposed to indoor air, which would establish whether unintentional microbial contamination of the test materials occurred during the experiments. Fungal growth on the front, back, and side surfaces of each material was monitored periodically over a period of two months. For both sets of experimental conditions, the growth rate was assessed using the ASTM mold index technique which gives a rating ranging from 0 (no growth) to 4 (>60% area coverage) based on visual

observations [Appendix 3 and its supporting document]. Another evaluation method developed in this study utilized digital image processing (ImageJ) to determine the mold coverage area as a function of incubation time. Mold growth was evaluated by determining the time until mold growth covered 50% of each material surface ($T_{50\%}$ - days) and by determining the growth rate (μ-%/day) observed on each material [Appendix 3].

4.3. Key Findings

4.3.1. Artificial Inoculation with A. niger

Both internal (from material components) and external nutrient sources (carbonaceous and nitrogenous nutrients and vitamins) were shown to influence mold growth. Visually, the fungal growth on non green gypsum board and ceiling tiles was evident at 3 weeks when the spores were inoculated with added nutrients. This result suggested that the *A. niger* spores utilized the additional simple molecules, such as dextrose and ammonium sulfate from the yeast base solution, to germinate and thrive. Also, these two materials contained organic matter, e.g. paper, which supports mold growth. Interestingly, the additional nutrient appeared to be exhausted after three weeks of incubation, since the density of mold growth on ceiling tiles and gypsum board did not appear to increase significantly after a few additional weeks of incubation. The light growth was possibly due to the high spore inoculum density and the small sample area inoculated, which in turn may have led to a limitation in nutrients to support further mold growth [46]. In contrast to the spores inoculated with a nutrient solution, no growth at three weeks was evident from the spores inoculated in sterile water that had no additional nutrients; however, after five more weeks of incubation, light growth was observed on these spots suggesting that these organic materials can support mold growth even in the absence of external nutrients.

Mold growth was also observed on the paperless drywall which does not contain the organic content necessary to support mold growth. After three weeks of incubation, significant mold growth was observed at spore concentrations ranging from 10 to10^5 spores/inoculated spot, if additional nutrients were provided to the material surfaces. This finding suggests that even if a building material itself does not favor growth, when soil or

other organic materials accumulate on the surface, the material can support vigorous mold growth. This conclusion is supported by the earlier findings by Chang et al. [47], in their study of *Penicilium* and *Aspergillus* growth on conventional materials, both species grew on new and used ceiling tiles**.** In fact, these investigators observed that used ceiling tiles were more susceptible to fungal growth than new materials, likely because of the additional nutrients provided by soiling and an increased hygroscopicity caused by the dust that settled on the material surfaces.

The remainder of the test materials (inorganic ceiling tiles, flooring materials, particle board and sunflower board) showed no visual growth even when the spores were inoculated with the yeast base, which contained all the additional nutrients and vitamins necessary for *A. niger* to grow. We suspect that this result is due to the quick penetration we observed of the spore suspension to the inside of the porous materials, such as with the inorganic ceiling tiles, immediately upon inoculation. Such penetration, however, may have allowed for fungal growth inside test materials, which would not have been detected from visual inspection of the surface in this study. In addition, for the inorganic ceiling tiles, the high pH environment [pH =11-13] that resulted when the ceiling tiles were wetted may have prevented mold growth: most fungi prefer an acid to neutral environment to thrive, e.g., pH in the range of 2.6-9.6 [48].

Spore concentrations and nutrient levels were also important factors that affected mold growth. Growth was often visible at the higher spore concentrations ($8x10^4$ spores/inoculated spot) whereas little or no growth was observed at the lower concentrations ($8x10^1$ spores/inoculated spot) during the first weeks of incubation. The study showed that it often took two additional weeks before mold growth was first observed for the lower spore inoculation levels. In this respect, the results with the gypsum board and ceiling tiles are typical examples. Also, mold growth on dry wall inoculated with spores in a yeast nitrogen base solution was only observed at the $8x10^4$ spore concentration, while growth was observed for all dilutions on the gypsum and conventional ceiling tiles. Another observation was that the 1X strength nutrient level supported less mold growth than did the 10X strength nutrient level. For example, substantial fungal mass was observed on the top surface of the ceiling tiles after three

weeks of incubation, while almost no growth was evident on ceiling tiles under the same experimental condition but with ten-fold fewer nutrients.

4.3.2. Natural Inoculation

Different surfaces (side, top and back) of the same material had quite different resistances to mold growth following natural inoculation. The differences may have been due to the different moisture and nutritional conditions impacting each surface. For instance, although it took almost two months for mold to cover 50% of the side surfaces of hardwood flooring, no growth was observed on its top and back surfaces over the same period (Figure 6). One possible explanation is that the side surface, which is more porous, absorbs more moisture and provides a larger growth subsurface for the mold. Another possibility is that the layer coating the top surface prevents the fungus from acquiring nutrients from the materials. An important effect of the coating layer was observed in the artificial inoculation experiments with *A. niger*. Following inoculation with a 10 uL drop, the solution still remained as a droplet on the top of the bamboo surface even after 2 months of incubation; the coating layer prevented penetration of the nutrients and spores and inhibited mold growth.

Moisture appeared to play a key role on mold growth rates on the building materials. It is pretty obvious that water-saturated materials enhanced mold growth. For example, the time ($T_{50\%}$) needed for mold to cover half the surface area of ceiling tiles incubated under high humidity conditions was twice that required when the ceiling tiles were water saturated. For most of the selected materials, it took more than 20 days for spores to germinate when the materials were only exposed to high humidity conditions and not directly saturated. This finding agreed with the experimental observation that by day 20, the moisture content of each material reached approximately 80% of its equivalent moisture content values (EMC) for a given air humidity level. It is quite clear that spore germination occurred only when moisture content reach a required level. Also, EMC values showed a strong positive correlation with the growth rates of mold on several test materials, e.g. non-green ceiling tiles, particle board, and sunflower board, indicating that high EMC may indicate high susceptibility to mold growth. Two materials, gypsum board and hardwood flooring, were an exception to this trend. Pasanen et al (2000) determined that the paper faces and gypsum bulk in gypsum board have

different nutrient availabilities and capacities for moisture sorption [43]. For instance, moisture is stored primarily in the bulk gypsum layer. In addition, the hardwood flooring consists of three different engineered wood layers which may have different capacities for storing moisture and nutrients. Thus, a bulk EMC measurement of an organic material composed of layers with significantly different properties may not correlate directly with mold growth rate.

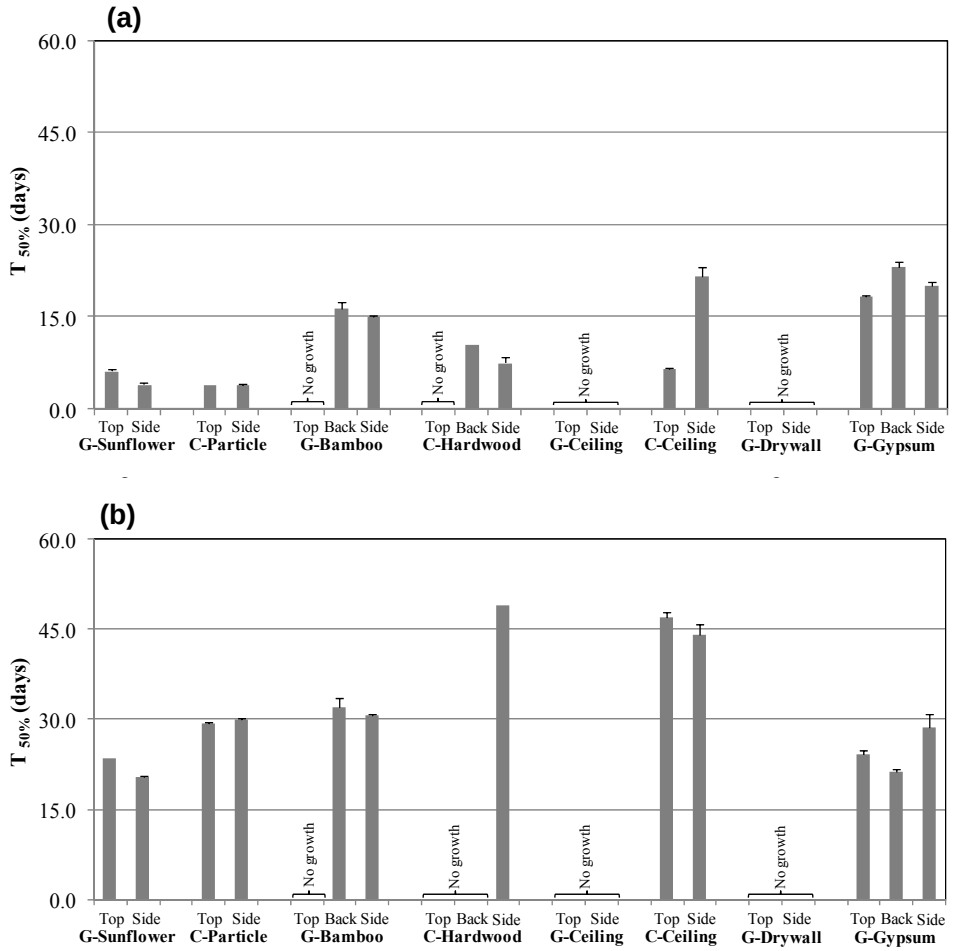


Figure 6. Time until 50% of the total material area was covered by mold following (a) direct water exposure or (b) high humidity exposure

Two different evaluation methods, the ASTM mold index and an image processing method of our own design, were used to evaluate the susceptibility of the test building materials to mold growth. The ASTM mold index approach was less time-consuming and was able to provide a quick and rough assessment of the extent of mold

growth on material surfaces. However, the ASTM mold index was somewhat ambiguous and quite dependent on an experimenter's judgment to decide which index should be applied for each growth case. Also it did not provide a quantitative assessment of the rates of mold growth that occurred – particularly at mold coverage percentages greater than 60%. In contrast, the image processing method was more time consuming but it provided a quantitative measure of the fungal growth rate (coverage percentage per day) that may be a helpful parameter for modeling fungal proliferation in indoor environments.

Possibly the most important conclusion resulting from this research section is that, based on our examination of a limited number of materials, the existing green rating does not necessarily warranty a material as being more resistant nor less prone to mold growth. Both test green materials that are organic based were quite susceptible to mold growth following natural inoculation. Under the same experimental conditions, sunflower boards or bamboo flooring obviously provided sufficient levels of nutrients to support fungal colonization. Other researchers have also observed that a range of fungal species can grow well on sunflower seeds [49-51] so growth on the sunflower board was expected. Nielsen et al (2004) evaluated growth of 16 fungal species on various building materials at 86% RH and 25^{o}C, and observed that approximately 50% of the area surface of beech wood and medium density fiber board was covered by mold after a 4 month period of incubation, whereas little growth was detected on gypsum [36]. Also, the cellulose-rich materials investigated in this study, such as particle and sunflower boards, yielded the greatest growth rate, as compared to other materials which were incubated under the same experimental conditions [Appendix 3].

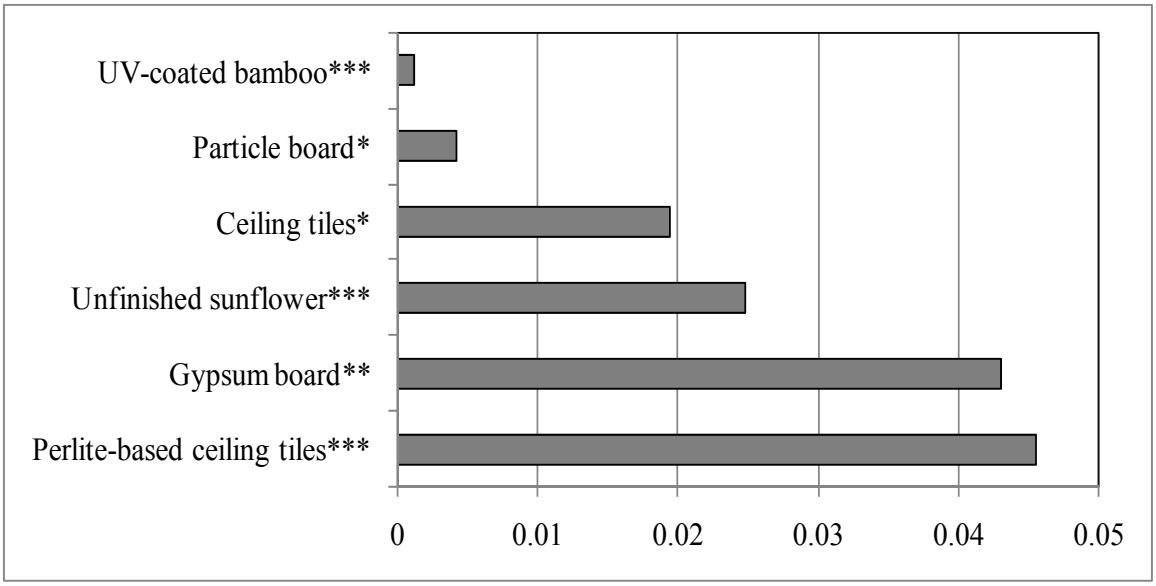
5. CROSS-COMPARISON AMONG TEST MATERIALS

Existing green product labeling programs for building materials are typically voluntary for the manufacturers and not yet supported by rigorous standards. It is inevitable that many of the criteria used to select green products are subjective, and a product may perform well under one criterion, but poorly under another. For instance, various sources, including this study, have demonstrated the fact that some building materials emit more VOCs after being exposed to ozone [2, 24, 31]. This finding suggests the importance of accounting not only for primary emissions, but also for secondary emissions when evaluating green materials. Thus, green materials that lead to lower secondary emissions will provide one more beneficial reason for using these materials. In addition, including the affinity of materials for fungal growth a rating criterion will provide direction for how to modify green materials to make them more resistant to mold growth.

This study involved two major phases: evaluating the reactivity of green building materials with ozone and determining their affinity for fungal growth. As discussed in Sections 1 and 2, three characteristics that are beneficial to building occupants and that should be considered in the selection of future green materials are:

1. Remove substantial amounts of pollutants (e.g. ozone) from air.
2. Emit little to no primary VOC or reaction products.
3. Be resistant to mold growth

These characteristics provide a means for comparing the "greenness" of the selected building materials evaluated in this research. For ozone removal, the values of deposition velocity reported in Figure 7 may be used to provide a relative rating; similarly, the total amount of VOCs emitted from selected materials following ozone exposure in Figure 5 may be used to rate the materials. Finally, the $T_{50\%}$ values reported in Figure 6 may be used to rate the relative mold resistance of each material. Table 4 shows the relative ratings ranging from 1 to 8 for each of the four pairs of green and non-green building materials tested in this research. A rating of 1 represents the best relative performance (highest ozone removal ability, lowest VOC emissions, and longest $T_{50\%}$ or highest mold resistance) while 8 is the worst.



* Poppendieck et al. 2007 [18]

** Grontoft and Raychaudhuri 2004 [52]

*** Hoang et al. [Appendix 1]

Figure 7. Mean deposition velocities of different materials

Table 4. Comparison of the "greenness" among test green and non-green materials

Materials	Ozone removal[1]		By-product formation[2]			Mold resistance[3]		
	V_d (cm/s)	Rank	TVOC (ng/l)		Rank	$T_{50\%}$ (days)		Rank
			Pre-exposure	Post-exposure		Top surface	Side surface	
*Perlite-based ceiling tiles**	0.046	**1**	5.47	1.73	**1**	N/D	N/D	**1**
Ceiling tiles	0.019	**4**	2.65	19.76	**4**	47.0	N/A	**5**
*UV-coated bamboo**	0.001	**6**	12.45	N/D	**2**	N/D	30.7	**4**
Hardwood flooring	N/A	**N/A**	13.67	3.50	**3**	N/D	49	**3**
*Unfinished sunflower**	0.025	**3**	48.46	26.20	**6**	23.4	20.4	**8**
Particle board	0.004	**5**	345.89	222.70	**8**	29.4	30.1	**6**
*Paperless drywall**	N/A	**N/A**	16.88	21.49	**5**	N/D	N/D	**1**
Gypsum board	0.043	**2**	21.85	76.66	**7**	24.2	28.7	**7**

*: Green building materials

[1]: rating based on the deposition velocity for non-green and green materials (Figure 7)

[2]: rating based on the total amount of VOCs emitted from selected materials (Figure 5)

[3]: rating based on the $T_{50\%}$ for direct water exposure (Figure 6)

N/A: not available; N/D: not detected

Among the selected materials, inorganic ceiling tiles rated the best with respect to ozone removal, minimal VOC emissions and resistance to mold growth. As discussed earlier, the inorganic composition and high alkalinity (pH = 11-13) of this material are likely responsible for the high fungal resistance of this inorganic ceiling tile. Interestingly, another non-organic material, paperless drywall board, which did not support mold growth by itself ranked number 1 along with inorganic ceiling tiles in the category of mold resistance (Table 4). However, mold growth was still observed on the drywall material with the support of an external nutrient source. This finding supports the observation that even new materials that do not initially support mold growth may support mold growth when soiled over time by dust and organic compounds. Also, paperless drywall did not emit much VOCs as compared to conventional gypsum board; however after being exposed to ozone, the paperless drywall emitted hexanal, octanal, nonanal, and decanal at a level which was not negligible.

Two other green materials did not rate as highly with respect to the three green criteria evaluated in this research (e.g., ozone removal, byproduct formation and mold resistance). The UV-coated bamboo flooring material did not remove as much ozone as the other cellulose-base materials did, although it emitted little VOCs even after ozone exposure. The coating layer appeared to protect reaction sites within the material from exposure to ozone. Also, the top surface of the bamboo flooring products did not support mold growth, likely due to the effect of a coating layer which prevented the mold from accessing the nutrients in the material. However, the side and back surfaces of bamboo flooring were quite susceptible to mold growth; thus they still rated poorly with respect to mold resistance. In addition to the significant susceptibility to mold growth, sunflower board also emitted a relatively large quantity of VOCs, although the emissions were much less than its non-green counterpart, particle board.

Although the green materials generally performed better than their non-green counterparts for the selected metrics (see Table 4), all of the materials, with the exception of inorganic ceiling tiles, performed well under one criterion but poorly under another. Thus, it would be valuable to carry out future work to investigate how to integrate these rating criteria in order to develop a more robust rating system for building materials. In addition, it is important to note that the described ratings are simply a rank ordering of

the performance of a given material relative to the other seven test materials examined in this study. This rating system of 1 (best performance) through 8 (worst performance) for the test materials must be interpreted with caution. For instance, gypsum board, which was rated second with respect to ozone reactivity, has a very similar deposition velocity to that of the first ranked material (perlite-based ceiling tiles). However, both perlite-based ceiling tiles and gypsum board had deposition velocity values which were nearly two times higher than those reported for the third and fourth ranked materials (unfinished sunflower and conventional ceiling tiles). Nevertheless, a rating system which includes relative metrics for assessing the chemical and biological reactivity of building materials represents a significant step toward the development of a more rigorous frame-work for rating green building materials.

6. CONCLUSIONS

The primary objective of this study was to facilitate the selection of green building materials, both explicitly and by defining a protocol by which future building materials can be evaluated. The study involved evaluating three different characteristics of building materials that can affect indoor air quality: (1) ozone removal, (2) by-product formation, and (3) fungal resistance.

To better understand the potential for ozone reactions with green building materials, ten common materials were exposed to ozone in stainless steel chambers. Deposition velocities and reaction probabilities for green building materials were of the same order of magnitude as their conventional counterparts. Key findings related to the ozone reactivity of green test materials are as follows:

- Inorganic ceiling tile may be a good choice for removal of ozone from indoor air. The ozone deposition velocity for ceiling tile was 0.05 cm/s, two times higher than wheat and sunflower board, and approximately ten times higher than ceramic and bamboo board.
- Coating layers appear to prevent ozone from reacting with material surfaces.
- Both transport and surface reaction resistances play key roles in ozone removal onto ceiling tile, while only surface reaction processes are important for ceramic tile, bamboo, UV-coated bamboo, and UV-coated wheat.
- Predictions of the ratio of indoor to outdoor ozone concentrations (I/O) suggest that green materials may not have a significant effect on indoor ozone concentrations, although the author acknowledges that more work is needed before definitive conclusions can be drawn on this issue.

The primary and secondary VOC emissions from four pairs of green/non-green materials (green/non-green ceiling tiles, sunflower/particle board, paperless drywall/gypsum board, and bamboo/hardwood flooring tiles) were also measured. Key findings related to the emissions of C_6 and greater carbonyls from reactions between test building materials and ozone are:

- Nonanal and decanal are common carbonyls emitted from building materials. Almost every test material contained nonanal and decanal and emitted these chemicals as

primary emissions. Particle board appeared to be the most significant source for C_6 to C_{10} carbonyls.

- The green materials tested in this study emitted less primary and secondary C_6 - C_{10} carbonyls than did their non-green counterparts. However, the difference was not statistically significant and the sample size precludes generalizations to all green building materials.
- Ozonation of materials may cause a reduction in the level of some chemicals emitted from selected materials. This might be explained by the reaction of the materials with ozone directly or with hydroxyl radicals which are formed by ozone reactions.

The results of the mold resistance experiments suggest that the green materials evaluated in this study were not necessarily more resistant, nor more prone to mold growth than were their non-green counterparts. Other key findings are:

- Cellulose-rich, organic-based, materials are more susceptible to mold growth than are inorganic materials for both green and non-green building materials. Following natural inoculation with indoor fungi, heavy fungal growth was observed on cellulose-rich materials, while paper-free materials, such as inorganic ceiling tiles and dry-wall, supported little to no growth.
- Direct wetting of materials expedites mold growth. The lag period until growth began was much shorter following direct water submersion than for the case where the materials were only subjected to high humidity conditions.
- A strong relationship was found to exist between equilibrium moisture content (EMC) and mold growth on naturally contaminated building materials. Cellulose-based materials with high EMC, such as sunflower and particle board, were especially susceptible to mold growth.
- Increasing spore levels and the presence of external nutrients promote the growth of fungi. Even materials such as paperless drywall which did not support growth in the absence of external nutrients, did support fungal growth when an external nutrient source was provided.
- Following natural inoculation, mold growth on the top surfaces and sides of a given material can be quite different, particularly for those building materials that have a top coating or consist of a heterogeneous composite of layered materials.

In conclusion, the three metrics, ozone removal, by-product formation, and fungal resistance, were selected to evaluate the performance of various green building materials. The overall results show that except for inorganic ceiling tiles, the other seven green and non-green materials performed well under one criterion, but poorly under another. Thus, future work on how to integrate the three selected metrics into a green rating system is worth consideration. In addition, there is certainly a need for further experimental and theoretical studies in order to fairly assess the benefits of the increasing use of green building materials, including:

- Measuring the formation of lighter carbonyls (C_5 and lower) from building materials exposed to ozone;
- Evaluating the role of hydroxyl radicals (OH*) chemistry following ozone reactions at indoor surfaces; and
- Studying the effects of coating layers and material composition on mold growth.

Additional fundamental studies that delineate the relationship between the composition of building materials and their chemical and biological reactivity would allow us to better predict the performance of both green and conventional building materials. Nevertheless, the research described in this dissertation provides the foundation for a more rigorous evaluation of green building materials, and hopefully motivates additional research in this important field.

APPENDIX 1 AND SUPPORTING DOCUMENT

Title: Ozone removal of green building materials.

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Ozone removal with green building materials

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Abstract

There is rapidly expanding market for green building materials. Such materials are intended to be environmentally friendly, with such characteristics as low toxicity, minimal chemical emissions, ability to be recycled, and durability. In addition, green materials often contain recycled and/or bio-based contents. Consequently, some green materials may undergo significant oxidation with potential for reduction of indoor ozone. In this study, 48-L electro-polished stainless steel chambers were used to study the reactive consumption of ozone by ten common green wall, flooring, ceiling, and cabinetry materials (perlite-based ceiling tile, unglazed ceramic tile, natural cork wall-covering, aluminum tinted cork wall-paper, bamboo, UV-coated bamboo, wheat board, UV-coated wheat board, sunflower board, and UV-coated sunflower board). Ozone removal was quantified in terms of deposition velocity and reaction probability. Ozone removal decreased with time after initial exposure, but for several materials the ability to react with ozone was regenerated after a period of zero ozone exposure. Test materials found to have the highest ozone reaction probabilities were a perlite-based ceiling tile, natural cork wall-covering, and wheat board.

Keywords

Indoor air; Green building materials; Ozone; Deposition velocity; Reaction probability.

1. Introduction

Indoor environments dominate total human exposure to many air pollutants due to the amount of time that most people spend indoors and the relative levels of indoor and

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outdoor air pollution. Zhang and Lioy [1] conducted a comprehensive study of ozone in residential air in New Jersey and found that indoor residential exposure alone accounts for 57% of the total exposure to ozone. Numerous other researchers have confirmed that the concentrations of volatile organic compounds (VOCs) and other pollutants are generally higher indoors than outdoors [2-4]. Also, 60% of total volatile organic compounds (TVOCs) in non-industrial buildings originate from building materials and furnishings [5]. Conventional building materials can produce formaldehyde, other toxic or irritating chemicals, and can react with ozone to produce secondary emissions [6-7].

Heterogeneous (surface) reactions between ozone and indoor materials are potentially important in terms of perceived indoor air quality [8-13]. Thus, it is important to understand the factors that influence reactions between ozone and indoor materials. The rate of ozone removal onto the surface of building materials is governed by a sequence of two steps:

- Transport of ozone to the material surface, which is dependent on the degree of mixing in core room air and the nature of the near-surface air flow.
- Ozone uptake onto the surface, defined by a reaction probability (γ), i.e., number of reactions of a molecule colliding with a surface divided by the total number of collisions.

The rate of ozone uptake by material surfaces has typically been quantified in terms of a deposition velocity or, and the reactivity of materials is typically quantified in terms of a reaction probability. Ozone deposition velocity onto materials is defined as the flux of ozone to a surface divided by its mean concentration in air:

$$v_d = \frac{J}{C_f} \tag{1}$$

where v_d is the ozone deposition velocity (m/s), J is the flux of ozone to a surface (mg/m^2/s), and C_f is the mean concentration of ozone in air (mg/m^3).

The inverse of deposition velocity is taken to be an overall resistance to heterogeneous reactions, and is equal to the sum of two process resistances in series:

$$\frac{1}{v_d} = r_o = r_t + r_s = \frac{1}{v_t} + \frac{1}{v_s} = \frac{1}{v_t} + \frac{4}{\gamma \langle v \rangle} \tag{2}$$

Where r_o is the overall resistance to removal at a surface (s/m), r_t is the transport resistance (s/m), r_s is the surface uptake resistance (s/m), v_t is the transport-limited deposition velocity (m/s), v_s is the reaction-limited deposition velocity (m/s), γ is the reaction probability (-), and $\langle v \rangle$ is the mean Boltzman velocity (= 360 m/s for ozone at 25^0C).

Equation 2 reflects both the effects of ozone transport to a surface and its interaction with that surface. If the transport-limited deposition velocity (v_t) is large and/or reaction limited deposition velocity (v_s) is small, the removal process should largely be influenced by reactions at the surface. This situation might occur, for example, in the presence of overhead fans and/or relatively low-reactivity materials. Conversely, if the transport-limited deposition velocity is small and/or the reaction-limited deposition velocity is large, the removal process should largely be influenced by the processes that affect ozone transport to a surface. An example might be a relatively "stagnant" room (little air motion) containing carpet or other fleecy materials that are highly reactive with ozone. Several authors have provided insightful discussions of the meanings of deposition velocity and reaction probability, and the relationship between the two [13-17].

Equation 2 can be rearranged to solve for reaction probability:

$$\gamma = \left[\frac{\langle v \rangle}{4} \left(\frac{1}{v_d} - \frac{1}{v_t} \right) \right]^{-1} \qquad (3)$$

The reaction probability is often estimated by determination of v_d and v_t through laboratory experiments. In these cases, v_d is determined first for a test specimen and then for the same or similar specimen that is chemically modified to substantially increase its reactivity. In the latter case, v_d is taken as v_t. The transport-limited deposition velocity is specific to the experimental system, while reaction probability, and hence v_s, should be specific to a given material.

Several researchers have reported deposition velocities and reaction probabilities for ozone and conventional (non-green) building materials. However, there is a paucity of published research related to ozone deposition onto green building materials (green materials), despite the fact that green materials are one of the most important components of green buildings.

There are several directories of green building products and materials to help customers and builders to choose green products [18-20]. However criteria used to select green products are subjective and a product may perform well under one criterion but poorly under another. One criterion that is sometimes used for green materials is that they are low emitting. This criterion generally applies to so-called "primary" emissions, typically of volatile organic compounds (VOCs), which are emitted from the actual components of the manufactured product. However, measurement of primary emissions of indoor materials alone may not be sufficient, since secondary emissions that are generated from ozone reactions with those materials may dominate over the time that a product is in use [21]. Understanding ozone reactivity with green materials is an important first step toward understanding the potential for secondary emissions from such materials, as well as determining green materials that effectively remove ozone without significant formation of by-products. In this paper, we focus only on the ozone reactivity of green materials. Deposition velocities and reaction probabilities are presented for ten green building materials.

2. Materials and Methods

2.1 Green Building Materials

The selection of green building materials was based on three main criteria:

- The materials are certified by a third party or listed on a well-established directory of green materials.
- The materials are prevalent in residential buildings and schools, and are commonly used for ceiling, flooring, wall-coverings, or cabinetry.
- The materials are available as both unfinished and finished products (tinted or UV-coated).

Ten green building materials were selected for this study: perlite-based ceiling tile, unglazed ceramic tile, natural cork wall-covering, aluminum-tinted cork wall-covering, bamboo, UV-coated bamboo, wheat board (pressed wheat straw), UV-coated wheat board, sunflower board (pressed husks of sunflower seeds), and UV-coated sunflower board. Some materials contained bio-based constituents (sunflower board, wheat board, cork wall-coverings, and bamboo), while some were inorganic, e.g., perlite-based ceiling

tile. The green ceiling tile consisted of 100% recycled and non-fibrous materials, including a blend of perlite and an inorganic binder. Ceramic tiles contained stoneware clays, minerals, and refractory contents with 50% recycled materials. Sunflower board and wheat board were produced from rapidly renewable resources, e.g., sunflower seed husks and wheat-straw, respectively. In addition, bamboo flooring was made from 100% rapidly renewable bamboo. Cork wall-coverings were produced from renewable cork oak.

All materials were unused when obtained; either shipped directly from manufacturers (ceiling tiles, ceramic tiles, and cork wall-coverings) or donated by a green builder in Austin, Texas (bamboo, wheat board, and sunflower board). Upon collection, materials were wrapped in multiple layers of plastic sheeting and stored for periods of up to several weeks before an experiment. Test materials and abbreviations used in this study are listed in Table 1.

2.2 Experimental System

A diagram of the experimental system is provided in Figure 1. Two 48-L electro-polished stainless steel chambers were operated in parallel. The two chambers were identical, with dimensions of 25 cm × 38 cm × 50 cm. After different materials were placed in each chamber, the chambers were sealed with face plates secured by a Viton ™ gasket and 20 bolts and wing nuts. A port on each face plate allowed air to be introduced to each chamber through a perforated Teflon tube that extended across the interior length of each chamber. Prior to each experiment, the chambers were cleaned with deionized water and dried with a heat gun in order to remove gases that had adsorbed onto, or particles deposited on, the chamber walls.

Laboratory air was pumped through a Perma Pure PD-series™ Nafion gas dryer to remove water vapor from the air, then through a UV light which converted oxygen into ozone. An activated carbon filter was installed after the UV light to reduce the ozone concentrations in the entering air stream to the desired concentration. The relative humidity of the supply air was adjusted by use of a split stream; one stream was passed through a water column (humidifier) before being reconnected to the main stream. A relative humidity probe (TSI, Inc., Q-TRAK™ model 8551) was installed to measure

both the temperature and relative humidity of the supply air. Two mass flow controllers (MFC) (Aalborg GCF171S) were used to maintain a constant air flow rate to each chamber. Ozone concentrations in the inlet and exhaust streams were determined by using single UV-cell ozone monitors (2B Technologies, model 202).

Each experiment was conducted under experimental conditions that are listed in Table 2. The mean inlet ozone concentration varied between experiments (100 ppb to 150 ppb). For the standard (base case) condition, temperature was maintained at between 22°C and 24°C, while relative humidity was controlled at 39% - 56%. The effects of a higher relative humidity of 70% were also studied for four green materials (natural/aluminum cork wall-coverings, ceiling tiles, and ceramic floor tiles). During each experiment, chambers were continuously ventilated with a flow rate of 800 ml/min (equivalent to an air exchange rate of 1 hr^{-1}). Each experiment was divided into three separate experimental phases. The first phase began when ozone was injected into the experimental system for a period of 48 hours. In the second phase, the UV lamp was switched off to discontinue the ozone supply to the chamber for a period of one day. After one day, the UV lamp was switched back on, and ozone was again injected into the chambers to assess whether the reactivity of the material had been regenerated.

To determine reaction probability, material samples were coated with a concentrated solution of sodium nitrite [1g of sodium nitrite ($NaNO_2$), 1g of potassium carbonate, 2 ml of glycerol in a solvent of 70 ml deionized water and 30 ml of methanol] and allowed to dry at room temperature. The same experimental procedure as described above was applied to $NaNO_2$-coated materials.

3. Data Analysis

If a chamber is operated as an ideal continuous-flow stirred-tank reactor (confirmed previously for experimental chambers), the concentration of ozone in the chamber is governed by the following equation:

$$\frac{dC}{dt} = \lambda C_o - \lambda C - v_d C \frac{A}{V} - v_{d,c} C \frac{(A_c - A)}{V} \qquad (4)$$

where C is the ozone concentration inside the chamber ($\mu g/m^3$), C_o is the ozone concentration entering the chamber ($\mu g/m^3$), λ is the air exchange rate of the chamber

(1/hr), A and A_c are the projected exposed areas of the material and the chamber walls, respectively (m^2), v_d and $v_{d,c}$ are the ozone deposition velocities associated with the material sample and with the chamber walls, respectively (m/hr), and V is the volume of the chamber minus the volume occupied by the material (m^3).

The time varying deposition velocity for each material was estimated by solving Equation 4 for v_d with analysis of consecutive pairs of ozone concentrations as shown in Equation 5:

$$v_d(t=t_{ave}) = \frac{2/\Delta t\left[C^n - C^{n-1}\right] + \lambda\left[C_0^n + C_o^{n+1} - \left(C^n + C^{n+1}\right)\right]}{A/V\left[C_o^n + C_o^{n+1}\right]} - v_{d,c}\left(t=t_{ave}\right)\frac{A_c}{A} \qquad (5)$$

Here, the superscript "n" corresponds to successive time steps, and all other variables are as described previously. The deposition velocity is taken at a time calculated as the mid-point (linear average, t_{ave}) between consecutive ozone measurements. The time increment (Δt) corresponds to the time between successive ozone measurements. To determine the ozone deposition velocity associated with the chamber walls ($v_{d,c}$), the same experimental procedure described in the previous section was carried out for both empty chambers.

4. Results

4.1 Ozone Deposition on Green Materials

Examples of ozone (O_3) concentration profiles for two types of wall-board specimens: aluminum tinted and natural cork wall-paper, and two empty electro-polished stainless steel chambers are shown in Figure 2. From approximately 48 to 72 hours the UV lamp used to generate ozone was switched off. The degree of O_3 removal by each material is clearly observed by the distance of each curve from the input concentration. At steady state, the ozone concentration in the outlet of each empty control chamber was approximately 86% of that entering the chambers. This loss of ozone indicates that it is necessary to account for O_3 loss by chamber walls when evaluating O_3 consumption by material surfaces. Nevertheless, cork wall-paper clearly reacted with and removed ozone; the total ozone removed by cork wall-paper in the experimental system was between 44% and 54%.

The time-dependent ozone deposition velocities for ten different green materials and electro-polished stainless steel are presented in Figure 3. Differences in deposition

velocities among these selected materials are obvious. As expected, the deposition velocity for ozone onto electro-polished stainless steel reaches a constant and relatively small value after the first five hours. The steady-state deposition velocity for stainless steel of 0.00027 cm/s is much lower than those reported by Kleno et al. [9] for stainless steel (0.007 cm/s) and hand polished stainless steel (0.01 cm/s). Differences in deposition velocities between different stainless steel materials is likely due to the effects of electro-polishing to remove reaction sites, and possibly the methods employed to handle and clean the steel, e.g., skin oils or airborne particles deposited on steel can react with ozone. Deposition velocities measured for ceiling tile, natural and aluminum-tinted cork wall boards, wheat board and sunflower board were much greater than the deposition velocities determined for the other green materials tested in this study.

For each material, the deposition velocity decreased rapidly during the first 10 h, before it leveled off to an approximately constant value. The deposition velocity of the ceiling tile, for example, was about 0.15 cm/s during the first hour, and decreased to a relatively constant value of 0.05 cm/s after 10 h. During the second exposure phase, a regeneration of ozone deposition velocity was observed, followed by a second decay in deposition velocity. Regeneration was particularly pronounced for ceiling tiles and sunflower board.

A rapid decay in deposition velocities was likely due to a consumption of sites that are highly reactive with ozone. These sites might be associated with the materials itself or with particles that deposit on, or gases that adsorb to, materials. The regeneration of reaction sites on test materials after 24 h without ozone exposure is important. Such regeneration was likely not due to deposition of particles or adsorption of gases onto the materials during the 24 h period without ozone exposure, since test materials were contained in experimental chamber with a conditioned inlet air stream as described above. Instead, it is conceivable that reaction sites on material surfaces were consumed during ozone exposure, thus establishing a concentration gradient between reactive molecules in the test material matrix and the surface of the material. This would induce molecular diffusion of such molecules to the surface, with effective surface replenishment (or regeneration) of reaction sites. During experiments the deposition velocity decays but levels off as the rate of diffusion to the surface approaches the rate of

consumption of reaction sites by ozone at material surfaces. This is a potentially important phenomenon given variations in outdoor ozone (a major source of indoor ozone) and intermittent usage of indoor sources of ozone, e.g. daytime use of laser printer and photocopy machines in buildings. Further research is needed to ascertain whether the regenerative capacity of green materials is sustained over long periods, e.g. years, of ozone cycling.

For the purpose of assessing ozone deposition velocities among different materials, and also for comparison to previous research, time-dependent deposition velocities were averaged for the last day of the first exposure phase when deposition onto test materials had approximately leveled off. The mean ozone deposition velocities of ten green materials are presented in Figure 4. Values taken from the published literature are also included for comparison.

The ozone deposition velocities between test materials differed by approximately a factor of 50, from 0.001cm/s for UV-coated bamboo to 0.046 cm/s for ceiling tile. Importantly, the deposition velocities of the same material, with and without a coating or pigment, were considerably different for wheat board, cork, and bamboo. Materials with coatings or pigments yielded lower ozone deposition velocities, presumably due to coverage (shielding) of reaction sites by a solid coating or pigment particles that are less reactive with ozone. Two cork wall-covering products, for example, had ozone deposition velocities of 0.026 cm/s (aluminum-tinted cork) and 0.037 cm/s (natural cork). This is similar to the trend observed for wheat board and UV-coated wheat board, and for bamboo and UV-coated bamboo.

4.2 Effects of Relative Humidity on Ozone Deposition

Four different materials were tested under two different relative humidity (RH) conditions: approximately 45% and 70%, as shown in Figure 5. Within the range tested, deposition velocity varied only slightly with variations in RH. This finding is in agreement with results reported by Reiss et al. [13] for wallpaper. They observed that RH variations in this range have no effect on ozone deposition onto vinyl and paper wallpaper. Weschler [6] also reported that the effects of relative humidity and temperature have relatively little impact on indoor ozone concentrations.

4.3 Reaction Probabilities

The deposition rate of ozone onto indoor surfaces is dependent on both mass transport processes and surface reactions. In order to compare the impacts of the transport and surface reaction processes on ozone deposition velocity, the ratios of transport limited resistance to total (transport and surface reaction) resistance are presented in Table 3. Ceramic tile, bamboo, UV-coated bamboo, aluminum tinted cork, and UV-coated wheat yield a resistance ratio of less than 0.1; surface reaction processes are dominant in terms of ozone deposition onto these materials. In contrast, the resistance ratios for ceiling tile, natural cork, wheat and sunflower board range from 0.3 to 0.5, indicating that both transport processes and surface reactions play key roles in ozone removal to these test materials. Ceiling tile had the largest reaction probability ($\approx 10^{-5}$), an order of magnitude or more greater than the reaction probabilities for bamboo, UV-coated bamboo, and ceramic materials (Figure 6).

5. Discussion

The deposition velocity is specific to each indoor environment (in terms of v_t), as well as to each material and pollutant (in terms of γ). Although there is little published literature on deposition velocities for green materials, various researchers have reported ozone deposition velocities for many conventional materials. Deposition velocities for ozone onto several conventional materials are presented in Equation 4

The reported deposition velocities of flooring materials range from 0.17 cm/s for carpets, to 0.007 cm/s for linoleum and 0.003 cm/s for oiled ash [9]. Values of deposition velocity determined in this study for ceramic tiles fell within this range. Similarly, bamboo had a deposition velocity of 0.001 cm/s, close to those reported for linoleum and oiled ash [9]. Sunflower board and wheat board, which are used for cabinetry and furniture, yielded average deposition velocities of 0.028 cm/s and 0.025 cm/s, respectively. These deposition velocities are comparable to those reported for plywood [22]. Also, the deposition velocities of both cork wall-covering products are similar to those reported for polyethylene sheets and two times lower than those reported for wall-paper [12,13,23].

Surface modifiers applied to materials tested in this study appear to have prevented the exposure of ozone to reaction sites. Deposition velocities for several coated or tinted surfaces were approximately one-half of those for unmodified surfaces. The effects of surface modifiers on ozone deposition reported in this study have also been observed for gypsum board [9]. The most reactive of conventional materials with ozone appears to be unpainted gypsum board, a wall/ceiling material, with a deposition velocity of 0.8 cm/s [9]. Painted gypsum board has lower deposition velocities, with reported values as low as 0.042 cm/s [9].

Several authors have reported reaction probabilities for ozone and conventional materials that can be used to compare with the results determined in this study. For example, Sabersky et al. [22] measured the ozone deposition velocity for plywood with one varnished side. Reaction probabilities ranged from 6×10^{-7} to 5×10^{-6}. Our reaction probabilities for sunflower board and UV-coated wheat board are within this reported range, while those for bamboo and UV-coated bamboo are much lower. The reaction probability of ozone onto glass has also been reported to be about 0.2×10^{-6} [12], which is comparable with the reaction probability of the ceramic tile tested for this study. Our results for ceiling tile yield a reaction probability of 10^{-5}, which is close to the value reported for conventional ceiling tiles by Poppendieck et al. [10].

Interestingly, modified surfaces, such as aluminum-tinted cork, UV-coated wheat board, and UV-coated sunflower board, exhibited higher transport limited deposition velocities, v_t. In each case, the raw (un-modified) surfaces appeared rougher to the naked eye and touch. The same observation was reported by Morrison et al. [15] for galvanized sheet metal and coated duct liner: the flat surface (galvanized sheet) yielded a higher transport limited deposition velocity. These results suggest the importance of near-surface fluid mechanics and viscous interactions between materials and adjacent air that affect transport processes to a material.

6. Conclusions

Green materials may cover a large percentage of indoor surfaces in the future. Thus, these products will likely be major contributors to ozone removal in buildings. To better understand the potential for ozone reactions with green building materials, ten

common green materials (ceiling tiles, unglazed ceramic tiles, natural cork wall-coverings, aluminum-tinted cork wall-coverings, bamboo, UV-coated bamboo, wheat board, UV-coated wheat board, sunflower and UV-coated sunflower board) were exposed to ozone in stainless steel chambers. Deposition velocities and reaction probabilities for green building materials appear to be of the same order of magnitude as their conventional counterparts. Key findings related to the ozone reactivity of green test materials are as follows:

- Perlite-based ceiling tile was the most reactive test material with ozone, followed by natural cork wall-covering. The ozone deposition velocity for ceiling tile was 0.05 cm/s, two times higher than wheat and sunflower board, ten times higher than ceramic and bamboo board, and approximately fifty times higher than UV-coated bamboo.
- Transport and surface reaction resistances play key roles in ozone removal onto ceiling tile, while only surface reaction processes are important for ceramic tile, bamboo, UV-coated bamboo, aluminum tinted cork, and UV-coated wheat.
- Materials with coatings or pigments yielded lower ozone deposition velocities and reaction probabilities, indicating that coatings prevent some ozone from reacting with underlying materials.
- Within reasonable variation in buildings, relative humidity has little effect on ozone deposition velocity to the green materials tested in this study.

There is certainly a need for further experimental and theoretical studies in order to fairly assess the benefits of the increasing use of green building materials. It is also relevant to study the formation of secondary emissions from ozone reactions with certain green materials, which our team is now undertaking as a follow-up to the experiments described in this paper. Further research on the reactivity of green materials will hopefully facilitate the selection of green building materials in the future. For example, it would be valuable to identify green materials that remove substantial amounts of ozone from air while forming little to no reaction products.

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Table 1. Test materials, designations, and applications

Material	Designation	Manufacturer/Model	Indoor application
Unglazed ceramic tiles	Ceramic	Fireclay Tile/Debris Series	Flooring
Perlite-based ceiling tiles	Ceiling	Chicago Metallic/Novum	Ceiling
Unfinished bamboo	Bamboo	Smith & Fong/Plyboo	Flooring and/or others
UV-coated bamboo	Bamboo_UV	Smith & Fong/Plyboo	Flooring and/or others
Unfinished sunflower	Sunflower	Environ Biocomposites/Dakota Burl	Cabinetry and/or furniture
UV-coated sunflower	Sunflower_UV	Environ Biocomposites/Dakota Burl	Cabinetry and/or furniture
Unfinished wheat	Wheat	Environ Biocomposites/Biofiber^{TM} Wheat	Cabinetry and/or furniture
UV-coated wheat	Wheat_UV	Environ Biocomposites/Biofiber^{TM} Wheat	Cabinetry and/or furniture
Natural cork	Cork_Na	Innovations, in Wall-coverings/Invironmentals	Wall covering
Aluminum tinted cork	Cork_Alum	Innovations, in Wall-coverings/Invironmentals	Wall covering

Table 2. Experimental conditions

Material	Inlet ozone concentration (ppb)	RH (%)	Temperature (^{o}C)
Ceramic	110 ± 0.8	39 - 50	22-23
Ceiling	120 ± 0.6	40 - 49	22-24
Bamboo	130 ± 3.4	46 - 52	22 -24
Bamboo_UV	116 ± 5.4	50 - 56	23-24
Sunflower	122 ± 0.8	40 - 51	23-24
Sunflower_UV	154 ± 1.0	49-53	22-24
Wheat	100 ± 0.3	50 - 55	22-23
Wheat_UV	124 ± 2.7	40 - 45	22-23
Cork Na	127 ± 1.5	45 - 47	22 -23
Cork_Alum	136 ± 1.6	49 - 52	23-25

Table 3. Deposition velocities and relative resistances for ozone removal to test materials

Material	Deposition velocity (cm/s)		Resistance (s/cm)		Resistance ratio $r_t/(r_s+r_t)$
Ceiling	v_d	0.046	r	22	0.500
	v_t	0.091	r_t	11	
	v_s	0.091	r_s	11	
Ceramic	v_d	0.004	r	255	0.054
	v_t	0.072	r_t	14	
	v_s	0.004	r_s	241	
Bamboo	v_d	0.003	r	296	0.051
	v_t	0.066	r	15	
	v_s	0.004	r_t	281	
Bamboo_UV	v_d	0.001	r_s	875	0.022
	v_t	0.053	r	19	
	v_s	0.001	r_t	856	
Sunflower	v_d	0.025	r_t	40	0.277
	v_t	0.090	r_s	11	
	v_s	0.034	r	29	
Sunflower_UV	v_d	0.021	r_t	47	0.190
	v_t	0.112	r_s	9	
	v_s	0.026	r	38	
Cork_Na	v_d	0.036	r_t	28	0.303
	v_t	0.118	r_s	8	
	v_s	0.051	r	20	
Cork_Alum	v_d	0.023	r_t	44	0.088
	v_t	0.259	r_s	4	
	v_s	0.025	r	40	
Wheat	v_d	0.028	r_t	35	0.391
	v_t	0.072	r_s	14	
	v_s	0.047	r	21	
Wheat_UV	v_d	0.011	r_s	89	0.094
	v_t	0.119	r	8	
	v_s	0.012	r	81	

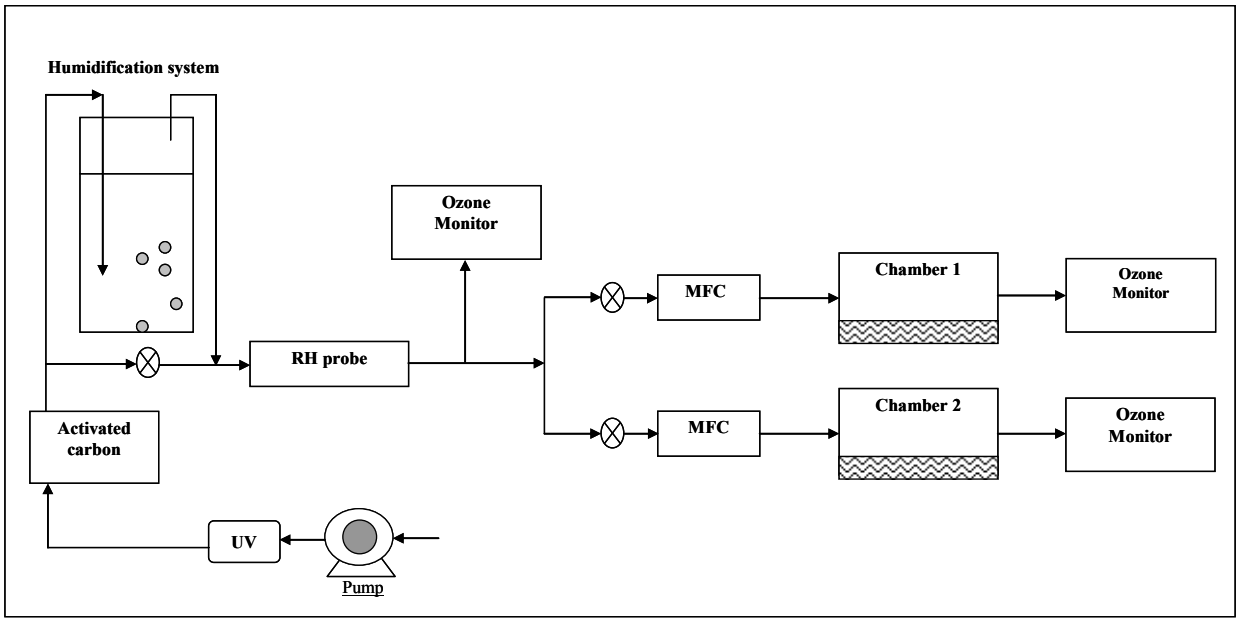

Figure 1. Experimental apparatus

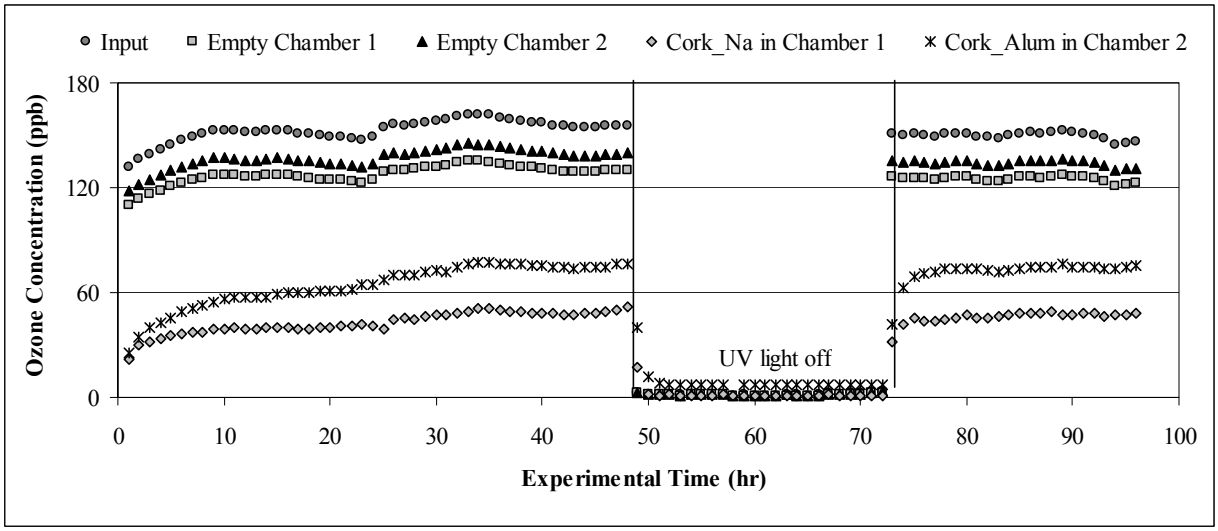

Figure 2. Ozone concentration profiles for empty chamber and cork wall-paper

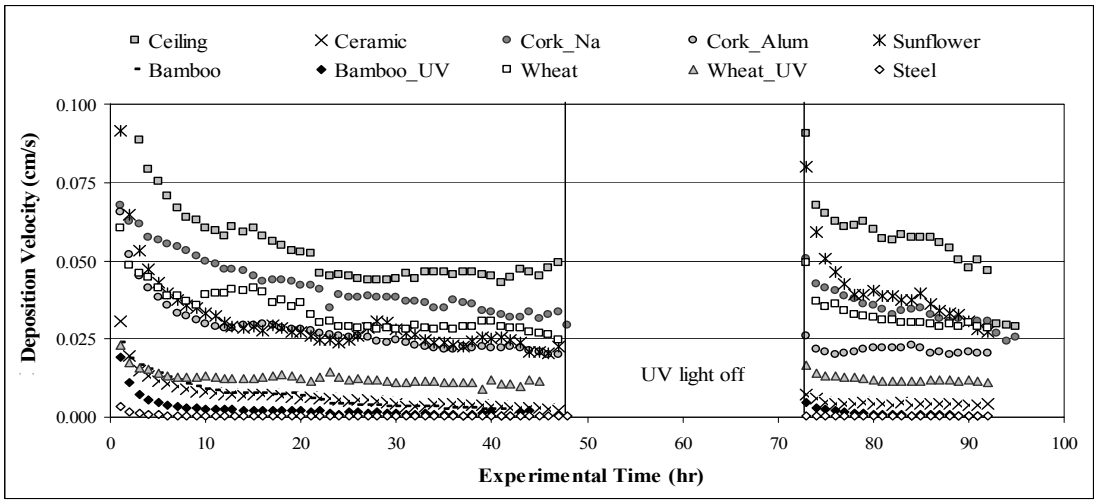

Figure 3. Time dependent deposition velocities for different green materials

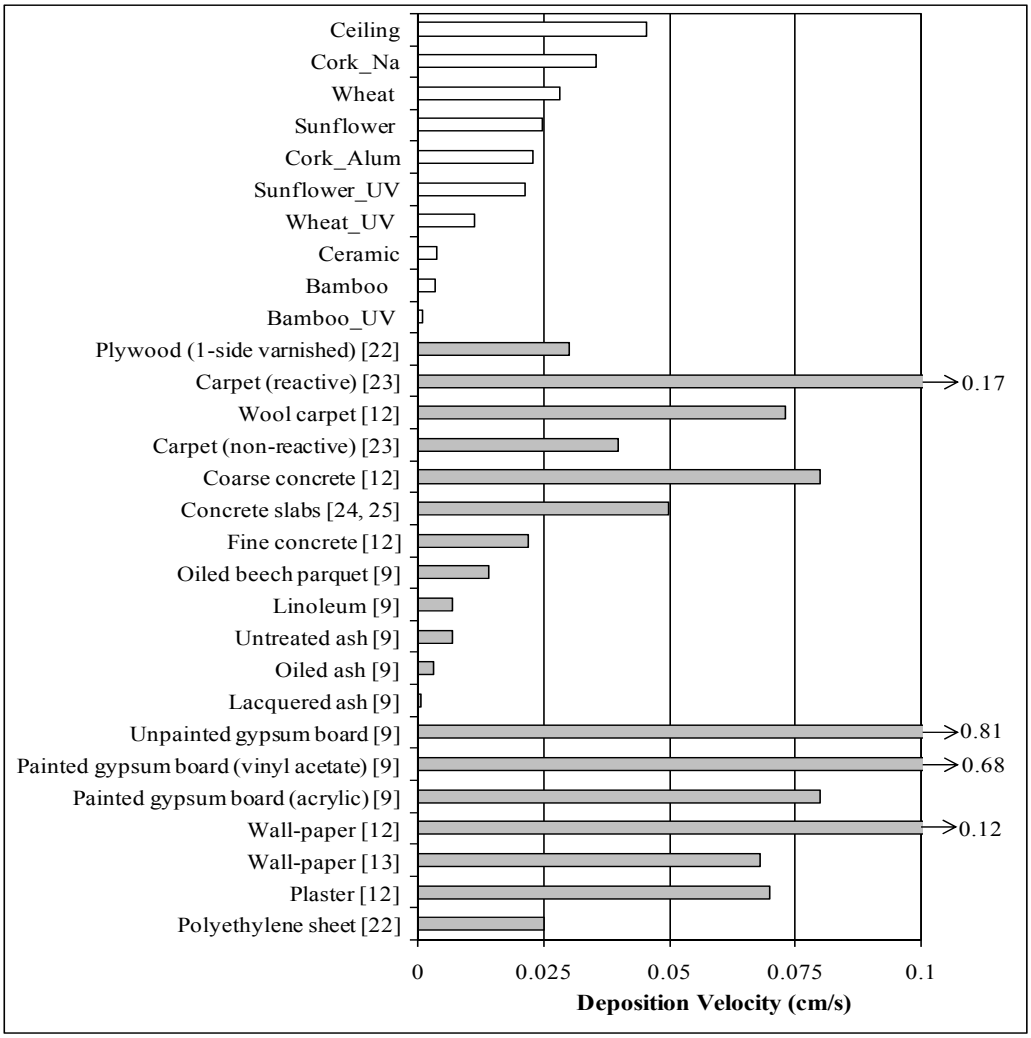

Figure 4. Mean deposition velocities of building materials

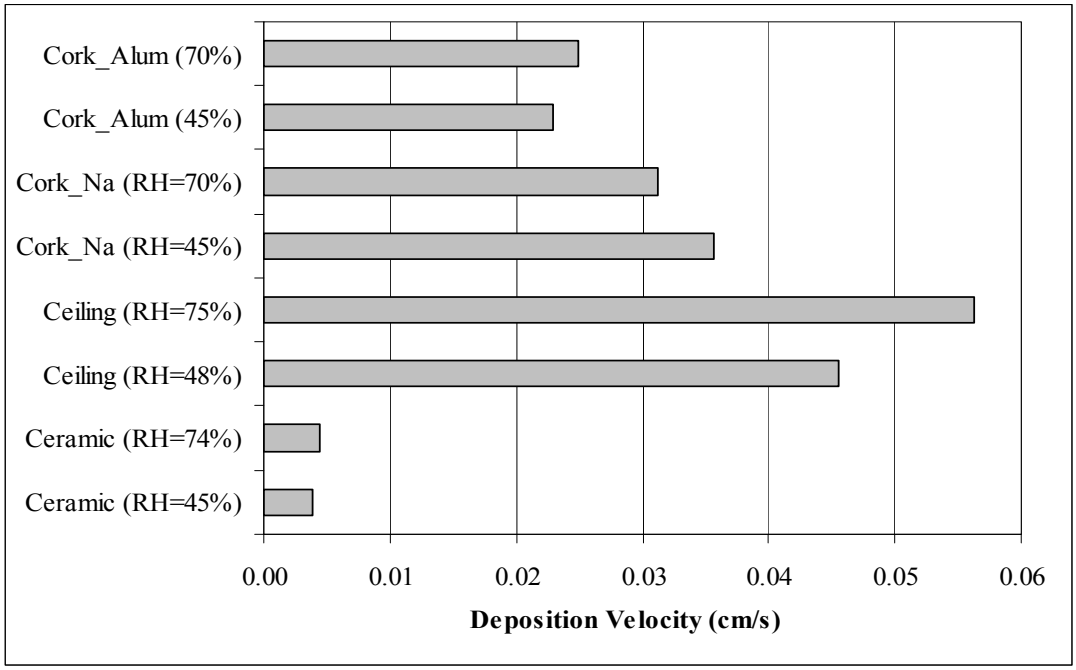

Figure 5. Deposition velocities at two different relative humidities

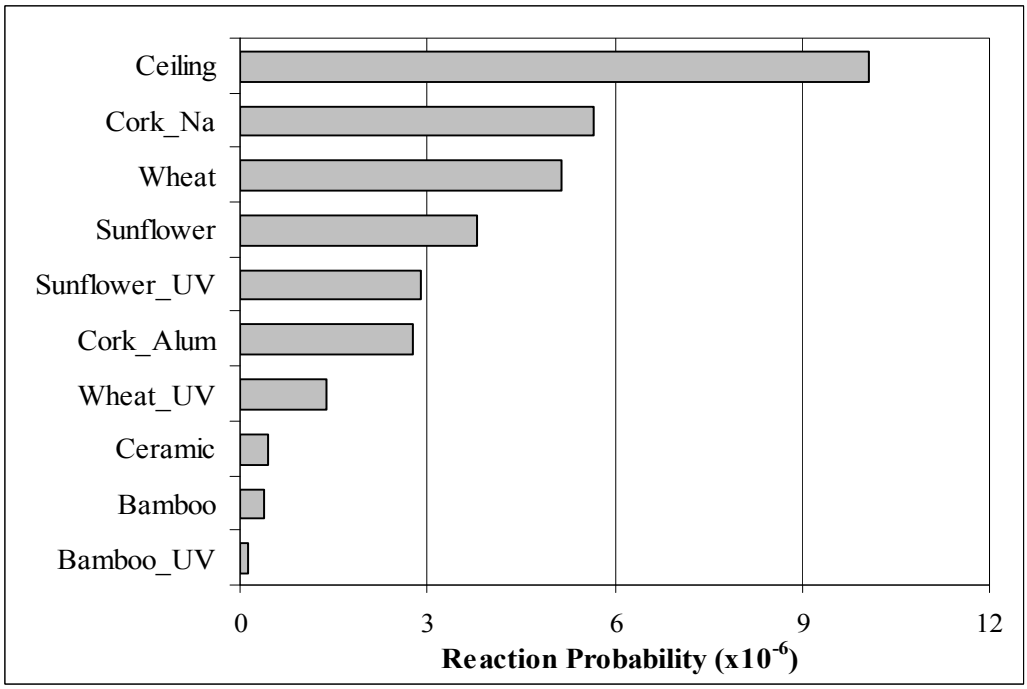

Figure 6. Reaction probability for green test materials

Supporting Document

Propagation of uncertainty

The time varying deposition velocity for each material was estimated by solving Equation 4 for v_d with analysis of consecutive pairs of ozone concentrations as shown as below:

$$v_d(t=t_{ave}) = \frac{{}^{2}\!/\!_{\Delta t}\left[C^n - C^{n-1}\right] + \lambda\left[C_0^n + C_o^{n+1} - \left(C^n + C^{n+1}\right)\right]}{{}^{A}\!/\!_{V}\left[C_o^n + C_o^{n+1}\right]} - v_{d,c}\left(t=t_{ave}\right)\frac{A_c}{A} \qquad (1)$$

where C is the ozone concentration inside the chamber (μg/m^3), C_o is the ozone concentration entering the chamber (μg/m^3), λ is the air exchange rate of the chamber (1/hr), A and A_c are the projected exposed area of the material and the chamber walls, respectively (m^2), v_d and $v_{d,c}$ are the ozone deposition velocities associated with the material sample and with the chamber walls, respectively (m/hr), and V is the volume of the chamber minus the volume occupied by the material (m^3). The superscript "n" corresponds to successive time steps, and all other variables are as described previously. The deposition velocity is taken at a time calculated as the mid-point (linear average, t_{ave}) between consecutive ozone measurements. The time increment (Δt) corresponds to the time between successive ozone measurements. To determine the ozone deposition velocity associated with the chamber walls ($v_{d,c}$), the same experimental procedure described in the previous section was carried out for both empty chambers.

$$Given\ X = C^n - C^{n-1};\ Y = C_o^n + C_o^{n+1};\ Z = C^n + C^{n+1}$$

$$Thus,\ \Delta X = \sqrt{\left(\Delta C^n\right)^2 + \left(\Delta C^{n+1}\right)^2}\ ;\ \Delta Y = \sqrt{\left(\Delta {C_o}^n\right)^2 + \left(\Delta {C_o}^{n+1}\right)^2};\ \Delta Z = \sqrt{\left(\Delta C^n\right)^2 + \left(\Delta C^{n+1}\right)^2}$$

We can rearrange Equation 1 as follows:

$$v_d = \frac{{}^{2}\!/\!_{\Delta t} * X + \lambda\left[Y - Z\right]}{{}^{A}\!/\!_{V} * Y} - v_{d,c}\frac{A_c}{A} \qquad (2)$$

The uncertainty for the deposition velocity (Δv_d) is was determined by using the following equation

$$\Delta v_d = \sqrt{\left(\frac{\partial v_d}{\partial X}\Delta X\right)^2 + \left(\frac{\partial v_d}{\partial Y}\Delta Y\right)^2 + \left(\frac{\partial v_d}{\partial Z}\Delta Z\right)^2 + \left(\frac{\partial v_d}{\partial v_{d,c}}\Delta v_{d,c}\right)^2}$$

$$Or\quad \Delta v_d = \sqrt{\left(\left[\frac{2}{\Delta t}\frac{V}{A}\frac{1}{Y}\right]\Delta X\right)^2 + \left(\left[-\frac{2}{\Delta t}\frac{V}{A}\frac{X}{Y^2} + \lambda\frac{V}{A}\frac{Z}{Y^2}\right]\Delta Y\right)^2 + \left(\left[\frac{V}{A}\frac{\lambda}{Y}\right]\Delta Z\right)^2 + \left(\frac{A_c}{A}\Delta v_{d,c}\right)^2}$$

Propagation of uncertainty for reaction probability

The reaction probability is often estimated by determination of v_d and v_t through laboratory experiments and can be expressed as follows:

$$\gamma = \left[\frac{\langle v \rangle}{4} \left(\frac{1}{v_d} - \frac{1}{v_t} \right) \right]^{-1}$$

Where v_t is the transport-limited deposition velocity (m/s), and $<v>$ is the mean Boltzman velocity (= 360 m/s for ozone at 25^0C).

$$\textit{Given} \quad X = \left(\left(\frac{1}{v_d} - \frac{1}{v_t} \right) \right) \textit{; thus} \quad \Delta X = \left[\left(\frac{1}{v_d^2} * \Delta v_d \right)^2 + \left(\frac{1}{v_t^2} \Delta v_t \right)^2 \right]^{0.5}$$

The uncertainty of the reaction probability ($\Delta\gamma$) is determined by using the following equation:

$$\Delta\gamma = \frac{4}{<v>} \frac{1}{X^2} \Delta X$$

APPENDIX 2 AND SUPPORTING DOCUMENT

Title: Ozone reactions with green building materials: Secondary emissions.

Authors: Chi P. Hoang, Kerry A. Kinney, Michael Waring, and Richard L. Corsi.

Journal: to be submitted soon as a technical note.

Ozone Reactions with green building materials: Secondary emissions

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Abstract

One criterion used to define a material as "green" is that it has low emissions of volatile organic compounds (VOCs). This criterion generally applies to primary emissions of VOCs that are emitted from the actual components of the manufactured product. However, many green materials are bio-based and consist of components that may be reactive with ozone, potentially resulting in secondary emissions of reaction products such as carbonyls. This study focused on emissions of C_6 to C_{10} saturated carbonyls from four different green building materials (bamboo, sunflower board, inorganic perlite-based ceiling tile, and paperless drywall) and four conventional (non-green) counterparts (hardwood flooring, particle board, cellulose-based ceiling tile, and gypsum board with paper) exposed to approximately 100 ppb of ozone. The green materials tested in this study emitted less primary and secondary carbonyls than did their non-green counterparts. However, the difference was not significant and the sample size precludes generalizations to all green building materials. Ozonation caused an increase in carbonyl formation for some materials (e.g. gypsum board and conventional ceiling tile). However some materials, such as particle board, exhibited a net reduction in emissions of carbonyls after ozonation, presumably due to oxidation of primary emissions to carboxylic acids or other reaction products that were not measured in this study.

Keywords

Secondary emissions, Ozone, Green building materials, Carbonyls

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1. Introduction

The use of green building materials in residential and commercial buildings has grown rapidly in the past several years. One often-used criterion for such materials is that they have low emissions of volatile organic compounds (VOCs) [1-2]. Wolkoff [3] classified emissions from building materials into two categories: primary and secondary emissions. Primary emissions refer to the release of free and non-bound VOCs from the material as manufactured. Secondary emissions include chemically and physically bound VOCs that are produced from interactions of materials with external parameters, such as heat, light, and reactive chemicals, e.g., ozone [4-8].

James and Yang (2005) studied primary emissions from three pairs of green and non-green materials, including Trex© versus pressure treated wood, ceramic versus vinyl composite floor tiles, and water-based versus oil-based paints [9]. The green materials emitted less VOCs than their non-green counterparts. However, ozone has been shown to react with building materials, such as gypsum board, latex paint, carpet and HVAC filters, to yield secondary by-product emissions [5,10,11]. Morrison and Nazaroff [5] explored secondary emissions from carpets and found an increase in some aldehydes, such as nonanal, after ozone exposure. Similarly, Reiss *et al.* [12] observed increases in emission rates of formic and acetic acid from latex paint when exposed to ozone (100-150 ppb). Secondary emissions from wood coatings were also observed as the products of ozone reactions with unsaturated acids (e.g., oleic and linoleic acids) and these emissions were observed to persist for years [13]. One recent study reported that ozone by-product emissions can persist for months or longer [14].

There is scant information related to ozone interactions with green building materials and corresponding secondary emissions of oxidized reaction products (ORPs). This technical note is intended as a follow-up to a previous paper we published related to ozone removal by ten green materials [15]. In this paper we explore the effects of ozone exposure on emissions from four green and four non-green building materials. We use a specific subset of C_6 and greater carbonyls for this screening analysis.

2. Experimental methodology

2.1. Test materials

Four pairs of green and non-green materials were selected for testing and are listed in Table 1, along with details related to material characteristics and composition. The green materials were all certified by a third party. The inorganic ceiling tiles were manufactured from expanded volcanic perlite, ceramic clay, sodium silicate, and an inorganic binder. According to the manufacturer, the product does not off-gas VOCs. The bamboo flooring was made from 100% rapidly renewable bamboo and low-emitting adhesives and coatings. The paperless drywall contained primarily continuous filament glass fibers, gypsum, and a small portion of silica crystalline. The sunflower board consisted of agricultural fiber and sunflower hulls.

Non-green ceiling tiles consisted of mineral fibers and up to 82% recycled content with high-performance engineering resin. The gypsum wall-board contained gypsum, cellulose, and starch. The gypsum core was encased in heavy natural-finish face paper on the front face and strong liner paper on the back face. The hardwood flooring was made of layers of wood stacked and glued under heat and pressure. The particle board contained 100% recycled ponderosa pine and was bonded with a resin. All materials were tested unused. The inorganic ceiling tile, paperless drywall, and bamboo flooring were shipped directly from manufacturers. The sunflower board was donated from a green builder in Austin, Texas. The four non-green materials were purchased from a home-improvement store in Austin, Texas. Upon receipt or purchase, materials were wrapped in multiple layers of plastic sheeting and stored for periods of up to nine months before an experiment. Material samples were cut into squares of 25 cm x 25 cm. prior to placement in an experimental chamber. To minimize emissions from, and ozone reactions with, the cut edges, the edge and bottom sides of each material specimen were coated with sodium silicate, an inert sealant.

2.2. Experimental conditions and plan

The experimental apparatus used for by-product formation experiments has been described in Hoang et al. [15]. Briefly, two 48 L electro-polished stainless steel chambers were operated in parallel, and each contained either a non-green or green material during

the same experimental run. Air was introduced to each chamber with an inlet ozone concentration of 120 to 140 ppb, temperature of 22-24^{o}C, and relative humidity of 40 to 55%. Chambers were continuously ventilated with an air exchange rate of 1 h^{-1}. Ozone concentrations in the inlet and exhaust streams were determined by UV absorbance.

2.3. By-product measurements

This screening study was focused on C_6 and greater carbonyls, which are easily collected on adsorbents and analyzed by thermal desorption followed by gas chromatography and mass spectrometry (TD/GC/MS). Based on previous research, twelve common indoor compounds were selected as target by-products: hexanal, 2-hexenal, heptanal, 2-heptenal, octanal, 2-octenal, nonanal, 2-nonenal, decanal, decenal, benzaldelhyde, and o-tolualdehyde (all were obtained from either Sigma–Aldrich or Fisher Scientific). Before each experiment, the experimental chambers were cleaned with de-ionized water and ethanol twice, and then heated at 205 °C for at least 24 h in order to eliminate chemical contamination between experiments. Following cleaning, the blank chambers were conditioned with filtered air for 8 to 10 h. Background samples were taken prior to beginning each experiment to ensure the chamber was clean.

Each experiment consisted of three-phases. Phase 1 (pre-exposure) involved the placement of a material in a test chamber for 8 h prior to exposure to ozone. Gas phase samples were taken to quantify the release of C_6 to C_{10} carbonyls associated with primary emissions from each material. During Phase 2 (ozone exposure), ozone was introduced to each chamber for 6 h. By-product measurements were not made during this phase. During Phase 3 (post exposure) ozone was no longer introduced in the inlet air. By-product samples were collected 4, 24, and 72 h after the termination of ozone injection in the chamber inlet stream.

By-products were sampled onto glass GC focus liners packed with 100 mg of Tenax-GR. Chamber air was collected at a constant flow rate of approximately 40 mL/min for 60 minutes. By-products were analyzed by TD/GC/MS, and quantified using an internal standard of 1-bromo-4-fluorobenzene (BFB). The experimental uncertainty in concentration measurements was 11-19% [16].

3. Results and Discussion

Carbonyl concentrations in the headspace of each chamber are presented in Figure 1. The graphs on the right show concentrations of C_6 and heavier carbonyls produced by green materials. Concentrations associated with non-green counterpart materials are shown on the left. Experiments were replicated and the results shown here are mean values. To assess the variation of carbonyl concentrations, the standard error is expressed as a symbol (┤) at the end of each bar. Among the twelve target compounds, only six aldehydes were detected: hexanal, heptanal, benzaldehyde, octanal, nonanal, and decanal. These carbonyls are also commonly found indoors. It is important to note that the concentration observed before ozonation correspond to primary emissions while those observed after ozonation correspond to a combination of both primary and secondary emissions.

Paperless drywall (G-Drywall) versus gypsum wallboard (C-Gypsum): Gypsum board and paperless drywall emitted similar amounts of hexanal during phase 1 (prior to exposure to ozone). Paperless drywall actually emitted more octanal than gypsum board did during phase 1. Gypsum board clearly emitted greater quantities of carbonyls following exposure to ozone, although the paperless drywall did emit slightly more hexanal and nonanal after ozone exposure than before. It is not clear as to whether the increased emissions from gypsum board stemmed from reactions with the paper surface or underlying gypsum. Poppendieck et al. [14] observed that gypsum board, especially gypsum backing, showed high deposition velocity, but had a relatively low by-product formation. This difference can be explained by the fact that gypsum products used in this study contained different additional components and composed of a fire-resistant gypsum core encased in heavy natural-finish face paper on the front face and strong liner paper on the back face. In this study, ozone may react with the front paper first, before it can penetrate further and react with gypsum. Paper has been observed previously to emit a significant amount of carbonyls following reaction with ozone [10]

For gypsum board, every carbonyl exhibited an increase in concentration 24 hours after ozonation before decreasing to a concentration similar to that observed prior to ozonation. The one exception was octanal, which was not observed prior to ozonation. A

similar observation was made by Poppendieck et al. [10, 14], the concentration of heavy carbonyls ($\geq C_8$) appeared to increase after ozone was flushed out of the chamber. Given their relatively low vapor pressures, e.g., 2 mm Hg at 20 °C for octanal and 0.26 mm Hg at 25 °C for nonanal, this continued release of carbonyls beyond the ozonation phase may be explained by their tendency to adsorb to materials and slowly desorb over time.

Sunflower (G-Sunflower) versus particle board (C-Particle): Particle board led to the highest carbonyl concentrations among eight test materials, and the total carbonyl concentration for particleboard was 10 times greater than the total carbonyl concentration for sunflower board. Hexanal was the greatest contributor to total carbonyl concentration for particle board, followed by benzaldehyde, octanal, and nonanal. A small amount of decanal was observed, but at a magnitude of one order of magnitude lower than the concentration of hexanal. Hexanal also contributed more than 80% of the total identified carbonyls for sunflower board. Relatively lower levels of octanal and nonanal were released after ozone exposure for sunflower board.

Compared to other selected green materials, sunflower board emitted the greatest amount of carbonyls. This observation is likely due to the fact that sunflower oils contain unsaturated fatty acids, such as oleic and linoleic acids, which can react with ozone leading to the formation of several oxygenated compounds such as hydroperoxides, ozonides, and aldehydes [7, 17]. An interesting observation is that for particleboard, and for hexanal associated with sunflower board, the concentration of each carbonyl was reduced significantly 4 hours after ozonation, and continued to decrease until the end of the sampling period. The concentration of hexanal associated with sunflower board dropped from 45 ng/L (before ozonation) to slightly lower than 20 ng/l (4 hours after ozonation), and then slowly decreased to about 13 ng/L at the end of sampling period. Similar observations have been reported by several other researchers [10, 14]. One possible reason for these observations is that ozone may react with compounds that were not quantified in this study and formed hydroxyl radicals (OH*), which then rapidly react with carbonyls associated with primary emissions to produce carboxylic acids and other carbonyls that were not quantified in this study. Pine-based particle board, for example, emits significant amount of α-pinene, which is known to react with ozone to form

hydroxyl radicals [18]. However, hydroxyl radicals are short-lived, such that scavenging by OH^* is likely decreased rapidly at the end of the ozonation period. The continued decrease in by-product formation 24 hours after the termination of ozonation requires further consideration.

Inorganic (G-Ceiling) versus non-green ceiling tiles (C-Ceiling): Four carbonyls (hexanal, octanal, nonanal, decanal) were observed to increase in concentration following the ozonation of non-green ceiling tile. The inorganic green ceiling tile emitted only small amounts of nonanal and decanal prior to ozonation, and secondary emissions did not appear to increase the concentrations of these two carbonyls after ozonation.

Bamboo (G-Bamboo) versus hardwood flooring (C-Hardwood): Both bamboo and hardwood flooring emitted small amounts of several carbonyls prior to ozonation. With the exception of hexanal emitted from hardwood, secondary emissions following ozonation did not increase concentrations of carbonyls for either material.

Summary

In this study we exposed four green and four non-green building materials to ozone and observed changes in C_6 and greater carbonyl concentrations in the chambers within which materials were placed. This rapid screening assessment suggests the following based on the test materials that we studied.

1. Paperless drywall emits less carbonyls than gypsum board containing paper surfaces, suggesting that ozone reactions with the paper on conventional gypsum board leads to secondary products.
2. Conventional particle board emits significantly more C_6 and greater carbonyls than sunflower board, both before and after exposure to ozone, suggesting that some green material alternatives to particleboard, e.g., for applications such as shelving, may reduce indoor concentrations of such compounds.
3. Inorganic (perlite-based) ceiling tile emits less C_6 and greater carbonyls than conventional ceiling tile. Recent experiments by our research team also indicate that the perlite-based ceiling tile is effective at removing ozone [15]. As such, this

material may be an attractive alternative for not only reducing primary and secondary emissions of carbonyls, but also for reducing indoor ozone concentrations.

4. Hardwood and bamboo flooring both emit only small amounts of C_6 and greater carbonyls.

The authors acknowledge the screening nature of the results presented herein. We only considered a small number of green materials and a subset of their possible counterparts. As such, it is impossible to develop generalizations of the indoor air quality benefits of green building materials based on this study alone. Only C_6 and greater carbonyls were studied. Future research should focus on lighter carbonyls and other oxidized products such as carboxylic acids. Finally, experiments were only completed to 72 hours after the termination of ozonation. The persistence of secondary products should be studied for longer time periods and with diurnal cycles of ozone exposure.

The authors hope that information provided in this technical note will serve as part of a growing base of information that engineers and architects can use in the future, and that this work motivates future research to better understand the role of primary and secondary emissions from green building materials.

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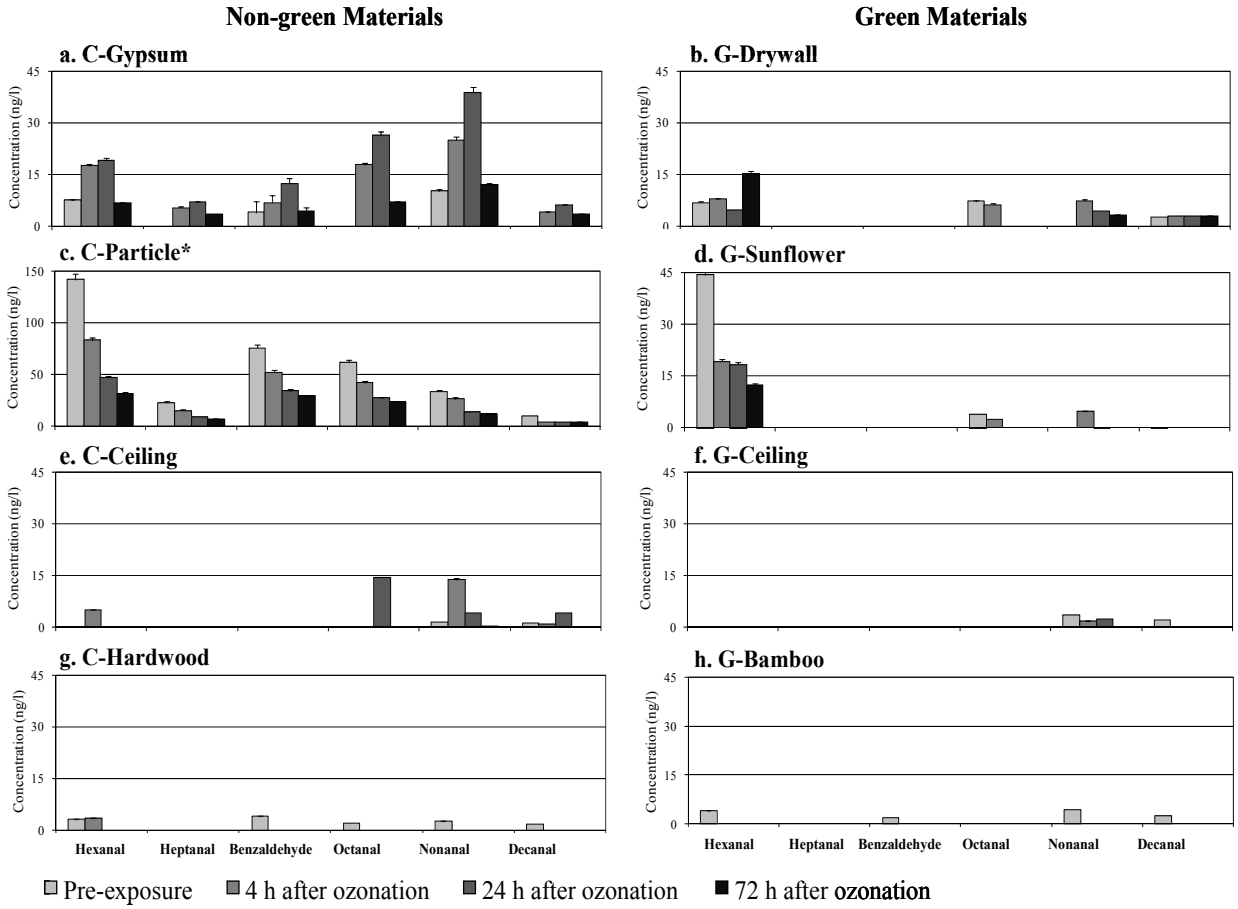
Table 1. Test materials, designations, and composition

Material	Indoor Application	Designation	Manufacturer/ Model	Composition	Low emitting labeled
Gypsum board	Wall covering	C[a]-Gypsum	USG/Sheetrock	Gypsum, cellulose and starch, fibrous glass (may be)	N/A
Paperless drywall	Wall covering	G[b]-Drywall	Geogria Pacific/	Gypsum, filament glass fibers, silica crystalline, quartz	N/A
Particle board	Cabinetry	C-Particle	Boise Cascade	Recycled ponderosa pine	N/A
Sunflower board	Cabinetry	G-Sunflower	Environ Biocomposites/ Dakota Burl	Agricultural fibers, sunflower hulls	Yes
Ceiling tiles	Ceiling	C-Ceiling	Armstrong/Homestyle	Mineral fiber, recycled content, resin	N/A
Inorganic ceiling tiles	Ceiling	G-Ceiling	Chicago Metallic/Novum	Perlite, ceramic clay, sodium silicate, and an inorganic binder	N/A
Harwood flooring	Flooring	C-Hardwood	Bruce/Homestyle	Oak, glue	N/A
UV-coated bamboo	Flooring	G-Bamboo	Smith & Fong/ Plyboo	Bamboo and low-emitting adhesives and coatings	Yes

[a]: Conventional
[b]: Green
N/A: Not available

Table 2. Experimental conditions

Material	Inlet ozone concentration (ppb)	RH (%)	Temperature (^{o}C)
G-Inorganic	125 ± 10	40 - 49	22 - 23
Ceiling	125 ± 10	40 - 49	22 - 23
G-Bamboo	141 ± 5.6	42 - 48	22 - 23
Harwood	141 ± 5.6	42 - 48	22 - 23
G-Sunflower	122 ± 3.6	50 - 53	23 - 24
Particle	122 ± 3.6	50 - 53	22 - 24
G-Drywall	130 ± 7.9	46 - 52	22 - 24
Gypsum	130 ± 7.9	46 - 52	22 - 24



* The concentration scale for particle boards is 0-150 ng/l while others are from 0-45 ng/l

Figure 1. By-product formation

Supporting Document

GC/MS for heavy carbonyl measurements

Gas samples for the analyses of individual carbonyls were collected from the center of the chamber through a Teflon exhaust line at a flow rate of 40 to 42 ml/min for 60 minutes using personal air sampling pumps. The detailed analytical procedure used in this study for carbonyl products was presented by and Waring (2008) [16]. In brief, sample air was captured on glass injection liners packed with 0.1 grams Tenax-TA^{TM}, then analyzed by thermal desorption gas chromatography/mass spectrometry (GC/MS) (HP5890 GC equipped with Atas Optic 2 thermal desorber and HP5971A mass selective detector), for a total run time of 21 minutes. A Restek Rtx 5SilMS capillary column, 30 m ID, 0.25mm internal diameter with film thickness of 0.5 um was used to separate organic compounds. The GC oven was held at the initial temperature of 40 °C for 2.5 minutes, after which it was ramped at 10 °C/min up to 150 °C and then 25 °C/min from 150 to 310 °C, at which it was held for 1.1 minutes until the end of the run time.

The response ratio for the target compounds were constructed using standards prepared from standard solution of carbonyls with dichloromethane. The stock solution (2µl of each target compound with 5 ml dichloromethane) was used to dilute to different mixtures of stock and di-chloro methane (1:1, 1:5, 1:10, 1:50). The relative response factors for each target compound relative to the appropriate internal standard BFB was calculated by using the following equation:

$$RR = \frac{A_x M_{BFB}}{A_{BFB} M_x}$$

where RR is the response factor (-); A_x, and A_{BFB} are area counts for the compound to be measured and BFB (internal standard) respectively; M_x and M_{BFB} are mass for the compound to be measured and BFB (ng).

The results of 3 to 8 successive samples were used to calculate the standard deviation. The relative standard deviations thus obtained were between 11% and 19%.

$$SD_{RR} = \sqrt{\sum_{i=1}^{n} \frac{\left(RR_i - \overline{RR}\right)^2}{n-1}}$$

where, SD_{RR} is the standard deviation of response factor; RR_i is the response factor at a concentration level *i*, $\overline{RR}$ is the mean response factor; and n is the number of concentration levels.

Determination of carbonyl concentration

The carbonyl concentration in chamber air was determined using the following euqation

$$C = \frac{M_x}{V_x}$$

where C is the compound concentration (ng/m^3); and V_x is the volume of air sampled (m^3)

Propagation of uncertainty

The uncertainty for the calculated compound concentrations (ΔC) was determined by using the following equation

$$\Delta C = \sqrt{\left(\frac{\Delta M}{V}\right)^2 + \left(\frac{M * \Delta V}{V^2}\right)^2}$$

Where Δ M is the standard deviation for compound mass (mg); and ΔV is the standard deviation for sampling air volume (m^3).

APPENDIX 3 AND SUPPORTING DOCUMENT

Title: Resistance of green building materials to mold growth.

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Journal: to be submitted soon.

Resistance of Green Building Materials to Mold Growth

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Abstract

While the market for "green" building materials for indoor uses has been expanding rapidly, the susceptibility of green materials to mold growth following moisture exposure is not well understood. The relative resistance of four pairs of green building materials (G-Sunflower board, G-Bamboo flooring, G-Ceiling tile, and G-Drywall wall-board), and their conventional analogs (C-Particle board, C-Hardwood flooring, C-Ceiling tile, and C-Gypsum wall-board) to surface fungal growth was assessed in laboratory experiments. An artificial inoculation protocol was used to investigate the effects of external nutrient levels, host material and spore levels on the susceptibility of eight building materials to a model fungal species (*Aspergillus niger*). A natural inoculation protocol was utilized to evaluate the resistance of the building materials to colonization by common indoor fungi after direct water exposure and/or following high humidity exposure. Increasing *A. niger* spore levels and the presence of external nutrients promoted the growth of *A. niger* on the surface of G-Drywall, C-Ceiling tile, and C-Gypsum wall-board. Following natural inoculation, cellulose-rich materials were found to be highly susceptible to mold growth after direct water exposure or following high humidity exposure. The time until 50% of the total surface area of a material specimen was covered by fungi ($T_{50\%}$), the lag period until growth began, and the fungal growth rates were useful metrics for comparing mold susceptibility among different building materials. Under the same experimental conditions, green perlite-based ceiling tiles (G-Ceiling tile), for instance, were found to be the least susceptible to fungal

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colonization while G-Sunflower board and C-Particle board were found to be the most susceptible. A strong correlation was found to exist between the equilibrium moisture content (EMC) of organic-based materials and $T_{50\%}$ for the top surface of these materials. Mold growth rates on the top, back and side surfaces of coated or composite building materials were quite different. Results suggest that the presence of organic matter in a given building material and its EMC are more important predictors of fungal susceptibility than is the label of "green" or "non-green."

Keywords

Green building materials; Fungal growth; Equilibrium moisture content; Mold

1. Introduction

Fungal proliferation inside buildings can detrimentally affect the health of building occupants (Nevalainen and Seuri, 2005) and cause discoloration and deterioration of building materials (Ezeonu et al., 1994; Klamer et al., 2004; Lee et al., 2006; Murtoniemi et al., 2003). Mold growth on conventional building materials is often linked to building dampness resulting from plumbing leaks, flooding or water condensation (Nevalainen and Seuri, 2005; Nielsen et al., 2004). Severe indoor mold growth is normally observed during wet periods, especially in humid areas. In some extreme cases, such as after Hurricanes Katrina and Rita in the Gulf Coast of the United States in 2005, heavy fungal growth developed in many building structures that remained flooded for weeks (Solomon et al., 2006; Rao et al., 2007).

A number of research studies have evaluated the effects of moisture level on the susceptibility of building materials to mold growth (Nevalainen and Seuri, 2005; Nielsen et al., 2004; Pasanen et al., 2000). When moisture levels are low, fungi generally do not grow on building materials or structures (Nevalainen and Seuri, 2005; Nielsen et al., 2004; Yang and Clausen, 2007; Black, 2006). A study by Pasanen et al. (2002) determined that relative humidity values ranging between 70% and 90% are required for fungal growth on building materials. They also observed that the relative humidity required for growth depended on the particular material and fungal species involved. Thus, moisture management is a critical component for controlling fungal growth on

construction materials. Moisture management requires an understanding of the water-holding capacity (WHC) and/or equilibrium moisture content (EMC) of a particular building material because both may be important indicators of the potential for fungal growth on a material exposed to water.

In addition to moisture, many other factors such as nutrient availability, spore levels, and fungal species affect mold growth (Murtoniemi et al., 2003; Nielsen et al., 2004; Pasanen et al., 2000; Yang and Clausen, 2007). For example, previous research has shown that fungal growth is dependent on the composition of the building materials. In particular, building materials such as ceiling tiles and wood furniture, which are organic or produced from organic products, can provide ample nutrients to support fungal growth (Black et al., 2006). Although fungi prefer sugars, amino acids and other simple nutrient forms, molds in nature generally acquire nutrients by breaking down more complex polymers such as starch, cellulose and lignin with the aid of extracellular enzymes (Black, 2006; Sedlbauer, 2002). Thus, a wide range of building materials is potentially suitable to support fungal growth. Antifungal additives such as sodium polyborate are often used to prevent fungal growth in susceptible materials (Black, 2006; Sedlbauer, 2002; Dillavou et al., 2007; Herrera, 2005); however, the short-term effectiveness of these additives and potential health concerns (Black, 2006; Nakayama et al., 2001) limit the popularity of anti-fungal additives for indoor uses. The preferred strategy is to select materials that are naturally resistant to mold growth and to eliminate the initial conditions that can lead to fungal growth (Rowan et al., 1999).

Building materials that are labeled “green” are becoming increasingly popular for in-home uses and are often touted as minimizing chemical emissions as well as being recyclable, and less toxic (Spiegel and Meadows, 1999). Although many conventional materials have been investigated to understand their susceptibility to fungal growth, there is a paucity of published research related to the affinity of green materials for fungal growth or to the effects of nutrient availability and spore levels on that growth. In addition, the question of whether green building materials are suitable for application in places that are subject to high humidity conditions or direct water exposure needs to be addressed. In order to provide guidance on the selection of building materials for moisture-sensitive indoor locations, this research focused on the following questions:

- How do external nutrient sources and fungal spore levels impact the growth of a model fungus on a range of building materials?
- How fast do mixed communities of fungi grow on the surfaces of green and conventional materials under high-humidity conditions or under nearly-saturated conditions?

To address these questions, the moisture uptake and fungal susceptibility of selected green materials utilized in flooring, ceiling tiles, wallboard, and cabinetry were evaluated and compared to their non-green counterparts. Two methodologies were employed to inoculate the materials: in one set of experiments, the materials were artificially inoculated with serial dilutions of *Aspergillus niger* (*A. niger*) spores suspended in a range of nutrient solutions; in the second set of experiments, the same materials were naturally inoculated by exposing them to an indoor environment containing a mixture of fungi.

2. Experimental methodology

2.1 Test materials

Four significantly different green materials were selected for this research study: inorganic sunflower board (G-Sunflower), bamboo flooring (G-Bamboo), ceiling tile (G-Ceiling), and paperless drywall (G-Drywall). These materials were listed as green in the directory of Greenspec and the Greenguard Environmental Institute. For comparison, a conventional counterpart for each green material was also tested: particle board (C-Particle), hardwood flooring (C-Hardwood), ceiling tile (C-Ceiling), and gypsum board (C-Gypsum) (Table 1). Two of the green materials, (G-Drywall and G-Ceiling) were made of inorganic materials, while the other two contained bio-based constituents. The four non-green materials contained wood (C-Hardwood and C-Particle) and/or some level of inorganic matter (C-Gypsum and C-Ceiling).

The eight building materials selected for study were new and unused. The green materials were shipped directly from manufacturers with the exception of the G-Sunflower material that was obtained from a green builder's workshop in Austin, TX. The four non-green materials were purchased from a national home improvement store located in Austin, TX. Upon collection, the materials were wrapped in multiple layers of

plastic sheeting and stored in a cardboard box for periods of up to several weeks before an experiment. The materials were cut to identical sizes (5 cm x 5 cm for moisture content and natural inoculation experiments and 3.8 cm x 3.8 cm for the water holding capacity experiments). Prior to use in the fungal growth experiments, the material specimens was sterilized by irradiation at a minimum dose of 25 kGy at the National Center for Electron Beam Research (Texas A&M University, College Station, TX, U.S.A).

2.2. Equilibrium moisture content

The measurement method for determining the equilibrium moisture content of test materials was adapted from the procedures described in ASTM D2216-05 (2005) and ASTM C1498-04a (2004). The building material specimens (5 cm x 5 cm) were first dried at 225°C until they reached constant mass. They were then sealed in 48 L, electro-polished stainless steel conditioning chambers maintained at a constant relative humidity (68% or 89%) and a temperature of 22-25°C. Air controlled at a relative humidity of 64-70% (or 85-92%) was passed through the chambers at a constant air exchange rate of 1 h^{-1}. The weights of the materials were determined periodically until they reached constant mass and the equilibrium moisture content of the materials was calculated as follows:

$$EMC = (M_{final} - M_{initial}) / M_{initial} x100 \quad (1)$$

where EMC is the equilibrium moisture content (%); $M_{initial}$ is the initial mass of dry material (g), and M_{final} is the mass of material at equilibrium with the water vapor in the chamber air (g). All measurements were made in triplicate and the values averaged.

2.3. Water-holding capacity

The water-holding capacity of each material was determined by submerging specimens (sized 3.8 cm x 3.8 cm) into water to mimic a flooding situation. Preliminary experiments indicated that 10 h was sufficient time for the material specimens to become water-saturated. The maximum water-holding capacity of the samples was expressed as the mass ratio of water to dry materials in Equation 2.

$$WHC = (M_{final} - M_{initial}) / M_{initial} x100 \quad (2)$$

Where WHC is the water-holding capacity (%); $M_{initial}$ is the initial mass of a dry material (g) and M_{final} is the mass of fully saturated material (g). All experiments were conducted in triplicate and the difference in mean values of WHC before and after rewetting were evaluated.

2.4. Artificial inoculation

The objective of this phase of the study was to compare the susceptibility of the test materials to colonization by a model fungal species in the presence and absence of additional nutrient sources, under ideal temperature and humidity conditions. *A. niger* was selected as the model fungus for this study due to its well-known association with indoor environments (Chang et al., 1995). The *A. niger* strain (ATCC 9642) was purchased from the American Type Culture Collection (Rockville, MD, USA). Preparation of spore inocula was adapted from the method described in ASTM G21-96 (2002).

Building material samples were systematically inoculated with serial dilutions of spores (conidia) of *A. niger.* The spores were diluted in either sterile-deionized water or with the following nutrient solutes supplied by Difco Scientific (Detroit, MI, USA): yeast nitrogen base (YNB), yeast carbon base (YCB) or yeast base (YB) in sterile water. The YNB contained all nutritive components (e.g. ammonium sulfate, amino acids and vitamins), except for a carbon source (Table 2). The YCB contained all nutritive components (e.g. dextrose, amino acids, and vitamins), except for a nitrogen source while the YB contained all nutritive components (e.g. ammonium sulfate, dextrose, amino acids and vitamins).

Experiment 1: 10X growth solution

To ensure that nutrients would not be limiting, the spore solutions used for the inoculations in this experiment contained a ten-fold (10X) higher concentration of nutrients than the level designed for liquid culture growth of fungi as recommended by the manufacturer (Table 2). Ten micro-liters of each spore solution, which was diluted to achieve 10^5, 10^3 or 10^1 spores for each inoculation spot, was applied by micropipette in duplicate to the surface of each building material tested. The non-inoculated area of the

material surface served as a negative control area. The inoculated samples were placed in Petri dishes, covered with a lid that allowed entry of air, and finally incubated in a conditioned chamber at a constant temperature of 30°C and a RH of 90-95% (controlled by a saturated K_2SO_4 solution). The extent of *A. niger* growth on each sample was visually evaluated and photographed (7.2 Mgp Panasonic Lumix DMC-LZ6 Camera) after three weeks and eight weeks of incubation.

Experiment 2: Standard growth solution

To evaluate the effect of lower nutrient levels, a 1X strength nutrient solution was used in this experiment to dilute the spores. Triplicate spots of $8x10^4$ or $8x10^1$ spores were applied by micropipette to the surface of each test material. The top and back surfaces of each material were tested separately. Spore-free solutions in sterile deionized water were also applied to the material's surfaces to serve as negative controls. In order to compare the resistance to mold growth among the test materials, the time until growth was first observed was determined with a 30X stereo microscope (Olympus SZ-ST, Olympus America Inc., Center Valley, PA, U.S.A) for each test material, growth solution, and spore concentration. The longer the elapsed time between the first observation of growth and the time of inoculation, the more resistant the material was considered to be to mold growth.

2.5. Natural inoculation

The purpose of this phase of the experiments was to investigate the growth rates of common fungi on the surfaces of both green materials and their non-green counterparts. The test was initiated by leaving previously sterilized building material samples (5 cm x 5 cm) unprotected in a residential house in Austin, TX, for ten days to allow for natural fungal inoculation. Because no additional specific fungal species was used to inoculate the material samples, this method was termed natural inoculation. Two different sets of experiments were carried out. In the direct water exposure experiments, the experimental set-up was designed to simulate what happens after a material is submerged in water (e.g., due to direct contact with floodwater or leaking water) and then, after the water drains away, is allowed to remain in a high humidity environment.

Specifically, after exposure to ambient air, the material specimens were first submerged in sterile water for ten hours to simulate direct exposure to water. The wetted samples were next placed in Petri dishes and incubated in a conditioning chamber as described earlier.

A second set of experiments was designed to investigate the mold resistance of indoor building materials exposed to high relative humidity conditions (approximately 90% or higher) for extended periods, a situation that can occur in a home built in a humid area. The experimental procedure was the same as that described above, with the exception that the materials were not submerged in water before incubation at a RH of 90-95%. For both sets of natural inoculation experiments, the area of fungal growth on the front, back, and side surfaces of each material was monitored periodically over a period of 2 months. Negative controls for both sets of experiments consisted of material samples submerged in sterile water for ten hours but not inoculated via extended exposure to indoor air. The mold growth rates after the materials were placed in the chambers were evaluated by determining the fraction of the material surface area that was contaminated by fungal mycelium.

An image processing tool was developed and implemented according to the three following steps: 1) identification of the area of fungal growth (see the red-bounded areas in Figure 1, for examples of such marked areas); 2) use of the ImageJ software package (Abramoff et al., 2004), developed by the U.S. National Institute of Health (NIH) to determine the surface area of the entire material sample (A_s - m^2) and that of the area contaminated with mold growth (A_c - m^2). The percentage of the surface covered by mold growth (P_c - %) was defined as $P_c=A_c/A_s x100$ and was determined as a function of incubation time for the top, bottom and side surfaces of each material sample. A rating of no or little growth (<10%) was confirmed by microscopic observation (30X) or with the aid of a digital image [taken by 7.2Mgp Panasonic Lumix DMC-LZ6 Camera].

In addition to determining the percentage of the surface area covered by mold growth (P_c - %) as a function of time for each material specimen, the fungal growth rate (μ - % day^{-1}), defined as the slope (dP_c/dt) of the exponential growth phase, was determined as described in Murtoniemi et al. (2003). Furthermore, to provide a metric to compare the mold resistance of the selected materials, the time until 50% of the total

surface area of a material specimen was covered by fungi ($T_{50\%}$, days) was also determined.

3. Results

3.1. Water uptake

The equilibrium moisture content (EMC) of the green materials at 64-70% and 85-92% relative humidity (RH) are presented in Figure 2. Sunflower board had the highest EMC values at both humidity levels, although those of the C-Particle, C-Hardwood, and G-Bamboo were only slightly lower. In contrast, the inorganic ceiling tiles (G-Ceiling) had a very low equilibrium moisture content of 0.7% at 64-70% RH and 2% at 85-92% RH. EMC values for the conventional ceiling tiles (C-Ceiling) were also relatively low, approximately 4% at 85-92% RH a value which is in agreement with a previous study conducted using four different ceiling tiles with EMCs ranging from 1% to 4% at 90% RH (Kumaran et al., 2006). The effect on the EMC of increasing the RH from 64-70% to 85-92% was greatest for sunflower board. After three weeks of incubation in the conditioning chambers, the eight test materials obtained moisture contents that were 80% or higher of their equilibrium values (data not shown).

The WHC values in Table 3 show significant variations among the test materials. Conventional ceiling tiles held the greatest amount of water (slightly more than 2.5 g water/g of dried materials), a finding of relevance given that ceiling tiles are often exposed to water from leaking roofs or pipes, or from condensation on pipes in open plenums above the tiles. In contrast, the bamboo flooring material presented the lowest water-holding capacity (approximately 12% or 0.12 g water/g of dried materials), which was one third that of the other organic products (C-Hardwood and G-Sunflower). The gypsum board, dry-wall, inorganic ceiling tile and particle board all had essentially the same water-holding capacity. Also, the data in Table 3 indicate that as little as 10 hours was sufficient time for the materials to become water-saturated. No statistically significant differences (at the $p = 0.05$ level) were detected in the mean values of WHC after the first 10-hour-period of water submersion and after rewetting. With the exception of the sunflower board, which tended to fall apart after being soaked, all of the other

material held their normal form and shape, even after being submersed in water for more than three days.

For several of the materials, the EMC and WHC values were quite different; for instance, the WHC of bamboo was smaller than that of the other materials while its EMC was relatively high. In contrast, the moisture uptake of the inorganic ceiling tile at RH = 85-92% was small (2%) whereas its WHC was nearly 1 g water per g of dried material. A similar finding was observed by Klamer et al. (2004) who measured the water-holding capacity of six different insulation materials and found that glass wool insulation which had a very high moisture uptake at room conditions had a low water-holding capacity.

3.2. Artificial inoculation

Following artificial inoculation with *A. niger* conidial suspensions in a 10X strength nutrient solution, the paperless dry wall (G-Drywall), ceiling (C-Ceiling) and gypsum board (C-Gypsum) materials exhibited visible fungal growth by week three of the incubation period (Figure 3); none of the other five material specimens developed growth on their top surfaces (data not shown). No obvious growth was produced at three weeks by the spores inoculated in sterile water that contained no additional nutrients; however, after five more weeks of incubation at high RH, light growth was observed on the ceiling tile (C-Ceiling) and gypsum board (C-Gypsum). For the materials that supported fungal growth, the spores inoculated with the yeast base solution yielded the heaviest growth after both three and eight weeks of incubation; this was true even for the paperless drywall material, which consists of inorganic materials. Also, less growth was observed when the spores were inoculated with the yeast nitrogen base, which contained all the required nutritive components and vitamins, but no carbon source.

When the artificial inoculation procedure was repeated with the 1X strength nutrient solution, again only the ceiling tile (C-Ceiling), gypsum board (C-Gypsum) and paperless drywall (G-Drywall) supported fungal growth during the eight weeks of incubation. The time, until mold growth was first observed at the $8x10^4$ and the $8x10^1$spore levels for each material, is summarized in Figure 4. The paperless drywall (G-drywall) showed heavy growth by the end of the first week of incubation (Figure 4b) when inoculated with additional nutrients. Subsequently growth was observed on the

back surfaces of the ceiling tiles and gypsum board. In several instances, the levels of mold growth were different on the back and the top surfaces of the same building material. For instance, after two weeks of incubation, all inoculated spots on the back of the conventional ceiling tiles (C-Ceiling) exhibited mold growth, whereas no growth was detected on their top surface until week six, except when the spores were inoculated with the yeast base solution (Figure 4).

3.3. Natural inoculation

Figure 5 presents the percentage of the material surface area covered by fungal growth as a function of incubation time following natural inoculation with a mixture of fungi. When material samples were subjected to direct water exposure prior to incubation, mold growth was observed within the first week of incubation for several materials (Figure 5a1 and 5b1). Sunflower board (G-Sunflower) appeared most suitable to support mold growth following water submersion. For example, after six days of incubation, substantial fungal biomass covered nearly the entire surface of the sunflower board. Extensive mold was also observed on the particle board (C-Particle) after one week. Fluffy mycelium grew heavily along the sides and top surfaces of the particle board, and black conidia were observed later during the incubation period (Figure 1). As is evident in Figure 5, after the spores started to germinate, the mold growth covered the materials very quickly. For some materials, such as C-Particle board, G-Sunflower board, and C-Gypsum, the entire surface area was covered by mold within one week after visible mold growth was first observed. In Figure 5a1, for example, the mold coverage across the sunflower board and particle board rapidly increased from 0% to 100% within 5 days. In contrast, most of the other materials needed more time to be covered by mold. This was especially true in the case of high humidity exposure, where, for instance, one-month of exposure was required for the ceiling tile (Figure 5c2) and even longer for the gypsum's side surface (Figure 5d2). Of note was the observation of mold growth on only 10% of the sides of the ceiling tile but on the entire top area after the first two weeks of incubation (Figure 5c2). In contrast, the opposite occurred with the bamboo and C-Hardwood flooring tiles. After two months of incubation, almost the entire coated surfaces of both flooring materials were free of mold, whereas all of the side areas were

covered heavily by a variety of obviously different mold species (Figure 5b1). Even after two months of incubation, no growth was observed on the paperless dry-wall (G-Drywall) and the inorganic ceiling materials (G-Ceiling) which do not contain a carbon source (Figure 5c1 and 5d1).

Essentially the same mold growth patterns were observed with the samples exposed to high humidity conditions (Figure 5c2 and 5d2). For example, sunflower and particle board were still the most favorable materials for mold growth. However, no growth was observed during the first three weeks of high humidity exposure suggesting that mold growth was retarded until the moisture content in each material reached a threshold level required to support growth.

In this study, we also introduce the parameter, $T_{50\%}$, (the time for common indoor fungi to cover 50% of the total material area) as a metric for comparing the mold resistances of different material samples under the same experimental conditions. Not unexpectedly, large variations were observed in the $T_{50\%}$ of the mycelium growing on different materials (Figure 6). Compared to the other materials, the mold growth on both cabinet materials (G-Sunflower and C-Particle board), again had the shortest $T_{50\%}$, (roughly 5 days) following direct water exposure, and 20 to 30 days if incubated instead under a high humidity exposure condition only. Figure 7 presents the correlation between the $T_{50\%}$ of the six organic-containing test materials and the EMC of these materials. With the exception of the top of the gypsum board (C-Gypsum) and the side of the hardwood flooring tiles (C-Hardwood) (see outliers in Figure 7), an increase in the EMC was linearly related to a decreasing $T_{50\%}$ ($R^2 = 0.95$).

The lag period and the maximum growth rate following natural inoculation of several test materials are summarized in Table 4. Both sunflower and particle board had the shortest lag time of three days following direct water exposure and about three weeks for the high humidity conditions. The growth rate for these two cabinet materials, varied from 20% to 50% per day depending on moisture conditions. Interestingly, the fungal growth rates on the materials exposed directly to water following natural inoculation was sometimes slower than that of materials exposed to high humidity following inoculation. We suspect that some spores that were naturally inoculated on material surfaces may

have been washed away during the submersion period; thus lowering the initial spore inoculation levels for the wetted material samples.

4. Discussion

Sedlbauer (2001) described the life cycle of a filamentous fungal colony as including two main processes: a vegetative growth phase (spore germination and hyphal growth) and a reproductive phase (sporulation). Whenever environmental conditions become favorable, viable spores of filamentous fungi start germinating and produce germ tubes, followed by the immediate production of a network of hyphae often referred to as a mycelium. After a certain amount of species-specific mycelial growth, spore formation may take place.

It is well known that fungi can cause health hazards and/or material damage during both their growth and reproduction periods (Nevalainen and Seuri, 2005; Chang et al., 1995). In general, mycelial growth leads to greater material building damage than do spores because the increase in mold coverage on material surfaces is mostly related to the mycelium using those materials or airborne organic dust (deposited on material surfaces) as nutrient sources. Figure 1 clearly shows the invasion of white mycelium across a particle board over time. Vasudeva (2004) has suggested that a concentration gradient develops around the mycelium, due to the utilization of nutrients in the growth region, which "drives" the fungal hyphae to grow towards the fresh substrate zone. The sporulation phase, however, is a major factor in public health because the spores, and less frequently hyphal fragments, released from fungal biomass growing on building materials can initiate allergic responses, toxic reactions and many types of infections in humans and other animals (Kildesø et al., 2003). Therefore, a material with less fungal mass (mainly due to mycelial growth), such as occurred with the hardwood flooring in the present study is still of concern, because such contaminated material may still release large quantities of unhealthy spores or hyphal fragments into the surrounding air.

Both internal (from material components) and external nutrient sources (carbonaceous and nitrogenous nutrients and vitamins) were shown to influence *A. niger* growth in the artificial inoculation experiments. Fungal growth was evident on C-Gypsum, C-Ceiling and G-Drywall at three weeks when the spores were inoculated with

added nutrients (Figure 4). This result suggested that the *A. niger* spores utilized the additional simple molecules, such as dextrose and ammonium sulfate from the yeast base solution, to germinate and thrive. Also, the two materials, C-Gypsum and C-Ceiling, contained organic matter, e.g. cellulose and starch, which can support mold growth. Considerably less growth was observed when the spores were inoculated with the yeast nitrogen base, which contained all the required nutritive components and vitamins, but no carbon source. These findings suggest that fungal growth may be carbon-limited on these particular building materials in the short term but not forever. Interestingly, the additional nutrients appeared to have been exhausted by three weeks of incubation, since the density of mold growth on ceiling tiles and gypsum board did not appear to increase significantly after a few additional weeks of incubation. It is possible that the light growth was due to the large spore inoculum size and the small sample area inoculated, which in turn led to the insufficient nutrient being available for mold growth (Vasudeva, 2004). In contrast to the spores inoculated with an additional nutrient solution, no growth at three weeks was evident from the spores inoculated in sterile water that had no additional nutrients; however, after five more weeks of incubation, light growth was observed on these spots suggesting that these organic-containing building materials can support mold growth even in the absence of external nutrients. The remainder of the test materials (G-Ceiling, G-Bamboo, C-Hardwood, G-Sunflower, and C-Particle) showed no visible growth even when the *A. niger* spores were inoculated with yeast base solution, which contained all the additional nutritive and vitamins necessary for *A. niger* growth. We suspect that this result is due to the quick absorption we observed of the spore suspension to the inside of porous materials, such as with the inorganic ceiling tiles immediately upon inoculation. Such penetration of the suspensions, however, could have allowed for fungal growth inside test materials, which would not have been detected from visual inspection of the surface in this study.

Spore concentrations and nutrient levels were also important factors that affected mold growth. Growth was often visible at the higher spore concentration ($8x10^4$ spores per inoculated spot) whereas little or no growth was observed at the lower concentration ($8x10^1$ spores per inoculated spot) during the first weeks of incubation. Our study showed that it often took two additional weeks before mold growth was first observed at the

lower spore inoculation levels. In this respect, the results with the gypsum board and ceiling tiles are typical examples (Figure 4a and 4b). Also, mold growth on dry wall inoculated with spores in a yeast nitrogen base solution was only observed at the $8x10^4$ spore concentration, whereas growth was observed for all dilutions on the gypsum and conventional ceiling tiles. Another observation was that the 1X strength nutrient level supported less mold growth than did the 10X strength nutrient level. For example, substantial fungal mycelium was observed on the top surface of the ceiling tiles after three weeks of incubation, while almost no growth was evident on ceiling tiles under the same experimental condition but with ten-fold less nutrients (Figure 4).

The effect of moisture on the growth rate of fungi on the materials studied was clearly evident. It is obvious that after direct water exposure, it took less time for mold to grow on the same materials. For example, the time required for mold to cover the 50% of the ceiling tile area ($T_{50\%}$) during high humidity exposure was twice that required for the sample exposed directly to water (Figure 6). For most of the selected materials, it took more than 20 days for spores to germinate after high humidity exposure (Figure 5, Table 4). This finding agrees with the observation that at day 20, the moisture contents of each material reached almost 80% of its maximum values (EMC) at the same air humidity level (85-95%). The results further suggest that spore germination occurred only after the moisture content of the materials reached a required threshold level. Although our study did not evaluate the effect of a range of ambient relative humidities on mold growth rate, a number of studies have observed that a longer time is needed for germination at lower relative humidities (RH) (Klamer et al., 2004; Nielsen et al., 2004; Pasanen et al., 2000; Adan, 1994). The increase in the observed *EMC* of selected materials in this study with increasing ambient RH (Figure 2) is in a agreement with the results reported by Kumaran et al. (2006) for eastern white pine, aerated concrete, and calcium silicate brick.

The mold growth rates on the cellulose-based materials showed a strong positive correlation with EMC (Figure 7). Thus, it appears that EMC values may be of use as an indicator for predicting the risk of mold growth on cellulose-based materials. Nevertheless, two exceptions were found for hardwood flooring tiles and gypsum board. As discussed by Pasanen et al. (2000), the paper face and the bulk gypsum phase present in gypsum board provide different nutrient contributions and capacities for moisture

sorption. Moisture, for instance, is stored primarily in the bulk gypsum layer. Similarly, the hardwood flooring consists of three different engineered wood layers, which may have different capacities for storing moisture. Interestingly enough, there was no correlation between WHC and the mold growth rates on the materials tested (data not shown). In fact, some materials with higher WHC seem less resistant to mold growth than those with lower WHC and vice versa. Ceiling tiles are a typical example.

Despite a range of experimental conditions, it was clear that the different surfaces (side, top and back) of the same material had quite different resistances to mold growth. The differences may have been due to the different moisture and nutritional conditions impacting each surface. For instance, although it took almost two months for mold to cover 50% of the side surfaces of hardwood flooring, no growth was observed on its top and back surfaces (Figure 6b). One possible explanation is that the side surface, which is more porous, absorbs more moisture and provides a larger growth subsurface for the mold. Another possibility is that the layer coating the top surface partially prevented the fungus from acquiring nutrients from the materials. An important effect of the coating layer was observed in the artificial inoculation experiments with *A. niger*. When the spore suspension solutions contained added nutrients, the solution still remained as a droplet on the top of the bamboo surface even after two months of incubation; but no mold growth was observed. This observation raised the question of whether the coating layer of the test materials contained natural or introduced antifungal properties that inhibited mold growth while the sides/back of those same materials, and presumably of their interiors, may still support such growth. This result suggests that future research on the effects of coating layers and material compositions on mold growth is desirable.

Possibly the most important conclusion resulting from this research is that, based on our examination of a limited number of materials, green materials were not necessarily more resistant, nor more prone, to mold growth than were their non-green counterparts. As is evident in Figures 5 and 6, both green and non-green materials that were organic based were quite susceptible to fungal growth following natural inoculation. The cellulose-rich green materials, sunflower board (Figure 5a1 and 5a2) and bamboo flooring (Figure 5b1 and 5b2) provided sufficient levels of nutrients to support fungal colonization. Other researchers have also observed that a range of fungal species can

grow well on sunflower seed hulls (Curvetto et al., 2002; Vuong, et al., 2004; Hasan, 1998), so growth on the sunflower board was expected. Surprisingly enough, these cabinet and flooring materials did not support growth of *A. niger* under any nutrient or spore level condition investigated. These results indicate that these materials may contain components which are not resistant to all fungal species, but are resistant to *A. niger*. Figure 6 also shows that it took only a short period of time for the cellulose-rich materials investigated in this study, such as particle and sunflower board, to be covered by mold in the natural inoculation experiments.

Not surprisingly, no growth was observed on inorganic ceiling tiles or paperless drywall inoculated naturally, even after two months of incubation, since these products contained no significant nutrients that would be expected to initially support mold growth. Also, the high pH environment [pH =11-13] that resulted when inorganic ceiling tiles were wetted may have prevented mold growth: most fungi prefer an acidic to neutral environment to thrive, and pH limits range from 2.6 - 9.6 (Carpenter, 1972). Karunasena et al. (2000) evaluated fungal growth on inorganic and cellulose-containing ceiling tiles and they also observed that the inorganic ceiling tiles did not support the growth of fungi of three selected genera (*Cladosporium, Penicillium, and Starchybotrys*) while growth of the same species was found on cellulose-containing ceiling tiles after only three days of incubation. However, *A. niger* in our experiments grew well on paperless drywall after three weeks of incubation if additional nutrients were provided to the material surfaces. This finding suggests that even if a building material itself does not favor growth, when soil or other organic material accumulates on the surface, the material can support vigorous mold growth (Murtoniemi et al., 2003). This conclusion is supported by earlier findings by Chang et al. obtained with *Penicilium* and *Aspergillus*, which were both shown to grow on new and used ceiling tiles (Chang et al., 1995). In fact, these investigators observed that the used ceiling tiles were more susceptible to fungal growth than the new materials, likely because of the additional nutrients provided by soiling and an increased hygroscopicity caused by the dust that settled on the material surfaces.

5. Conclusions

The study results suggest that the green materials evaluated in this study were not

necessarily more resistant, nor more prone to mold growth than were their non-green counterparts. Other key findings are:

- Cellulose-rich, organic-based, materials are more susceptible to mold growth than are inorganic materials for both green and non green building materials. Following natural inoculation with indoor fungi, heavy fungal growth was observed on cellulose-rich materials, while paper-free materials, such as inorganic ceiling tiles and dry-wall, supported no or little growth. The lag period until growth began was much shorter following direct water submersion than for the case where the materials were only subjected to high humidity conditions.
- Increasing spore levels and the presence of external nutrients promote the growth of fungi on building materials. Even materials such as paperless drywall, which did not support growth in the absence of external nutrients, can support fungal growth when an external nutrient source is provided. This finding suggests that new materials that do not support mold growth initially may support mold growth over time as they become soiled by dust and organic compounds.
- Following natural inoculation, mold growth on the top surfaces and sides of a given material can be quite different, particularly for building materials that are coated or are a heterogeneous composite of a variety of materials.
- Image analysis of fungal growth on material surfaces allows determination of $T_{50\%}$ and the fungal growth rate (in coverage percentage/time units) which are useful metrics for comparing the susceptibility of different building materials to mold growth following exposure to moisture. These parameters should be considered for inclusion in future rating systems for green building materials.

There is certainly a need for further studies to evaluate a broader range of green and non-green building materials to fairly assess the fungal resistance of these building materials. It is desirable, for example, to better understand the effects of material composition on mold growth. Nevertheless, this study provides useful metrics for comparing the mold susceptibility of green and conventional materials, e.g. the time until 50% of the total surface area of a material specimen was covered by fungi ($T_{50\%}$) or growth rate (μ). In addition, it is clear that the organic content and equilibrium moisture

content are important predictors for fungal susceptibility of building materials. The authors hope that the results provided herein motivate additional research related to mold growth on green building materials.

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Table 1. Characteristics of test building materials

Indoor Application	Material	Designation	Composition
Cabinetry	Sunflower board	G[a]-Sunflower	Agricultural fibers, sunflower hulls
Cabinetry	Particle board	C[b]-Particle	Recycled ponderosa pine
Flooring	Bamboo flooring	G-Bamboo	UV-coated bamboo and low-emitting adhesives
Flooring	Harwood flooring	C-Hardwood	Oak, glue
Ceiling[c]	Inorganic ceiling tiles	G-Ceiling	Perlite, ceramic clay, sodium silicate, and an inorganic binder
Ceiling[c]	Ceiling tiles	C-Ceiling	Mineral fiber, recycled content, resin
Wall covering	Paperless drywall	G-Drywall	Gypsum, filament glass fibers, silica crystalline, quartz
Wall covering	Gypsum board	C-Gypsum	Gypsum, cellulose and starch, fibrous glass

[a]: Green
[b]: Conventional

Table 2. Formulate for 1X strength nutrient solution[1] per liter

Nutrients	Yeast base	Yeast carbon base	Yeast nitrogen base
Nitrogen source			
Ammonium sulfate	5 g		5g
Carbon source			
Dextrose	10 g	10 g	
Amino acids			
L-histidine mono-hydrochloride	10 mg	10 mg	10 mg
LD-methionine	20 mg	20 mg	20 mg
LD-trytophan	20mg	20mg	20mg
Vitamins and trace minerals	*as specified by the manufacturer (Bacto-Difco)*		

[1]: 10X strength contains ten fold higher concentration of nutrients

Table 3. Water holding capacity of test materials

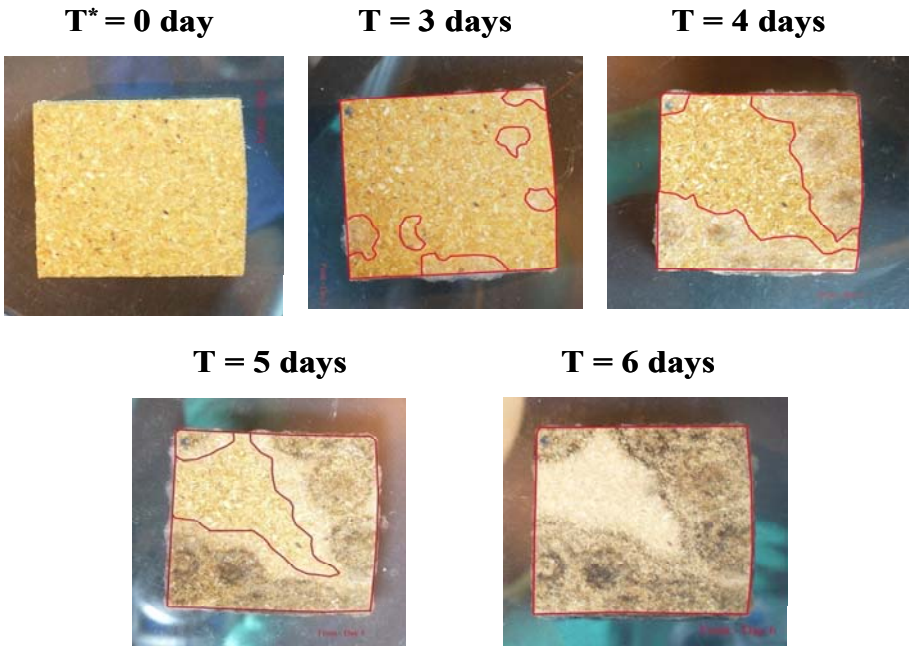
Materials	WHC (after 10 hrs) %	WHC (after rewetting) %	T-test P-values
G-Sunflower	31.25 ± 3.18	34.19 ± 3.39	0.17
C-Particle	82.63 ± 12.2	80.32 ± 14.69	0.42
G-Bamboo	12.35 ± 2.65	11.93 ± 2.26	0.42
C-Hardwood	31.98 ± 0.92	33.72 ± 1.92	0.12
G-Ceiling	79.89 ± 3.79	86.66 ± 7.04	0.11
C-Ceiling	255.08 ± 2.02	260.39 ± 8.13	0.17
G-Dry wall	78.85 ± 7.72	80.23 ± 11.84	0.44
C-Gypsum	83.27 ± 1.42	82.87 ± 3.54	0.43

Results are given as the mean value ± standard deviation.
*Indicate statistically significant difference between WHC before and after rewetting

Table 4. Mold growth rate on side surface of selected materials following natural inoculation in an indoor environment

Materials	Direct water exposure		High humidity exposure	
	Lag (days)	*μ (%/day)*	*Lag (days)*	*μ (%/day)*
G-Bamboo	12.3 ± 0.6	18 ± 2.5	28.0 ± 1.0	38.2 ± 5.4
C-Hardwood	5.7 ± 0.6	15.7 ± 4.0	32 ± 2.6	3.2 ± 0.2
G-Sunflower	2.7 ± 0.6	33.6 ± 4.1	19.7 ± 0.6	45.4 ± 7.3
C-Particle	3.3 ± 0.6	28.8 ± 3.4	26.3 ± 0.6	22.5 ± 4.1
C-Ceiling	17.3 ± 2.9	10.0 ± 2.2	26.7 ± 1.2	2.4 ± 0.2

Results are given as the mean value ± standard deviation.



*: Inoculation time

Figure 1. Determination of mold coverage area for water-submerged C-Particle using image processing method.

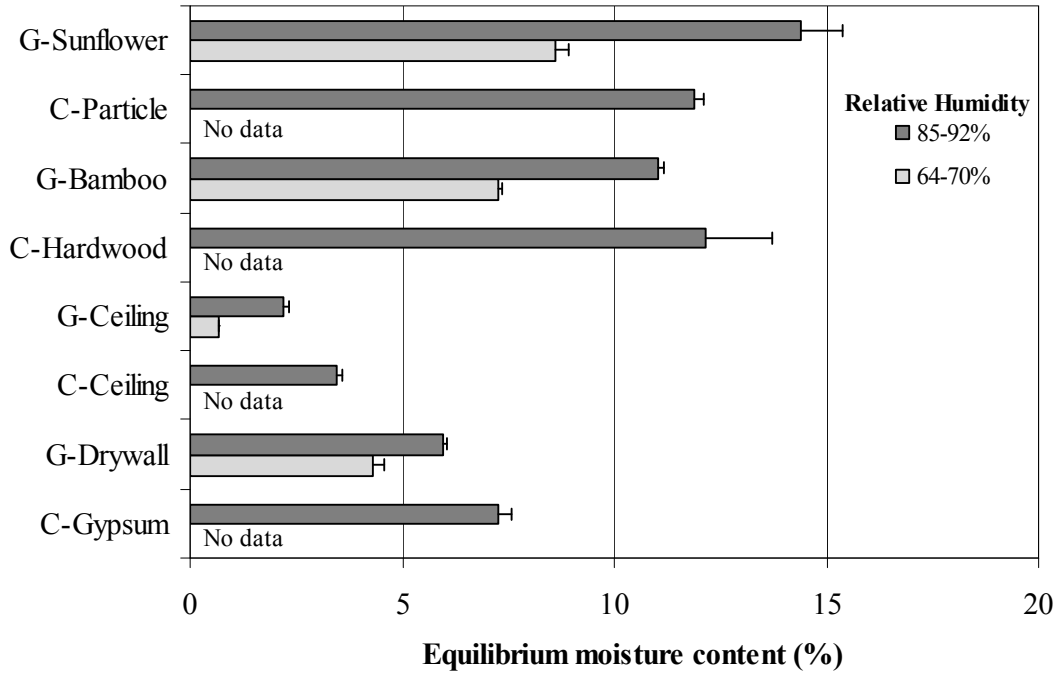


Figure 2. Equilibrium moisture content of green and non-green building materials. Error bars represent one standard deviation

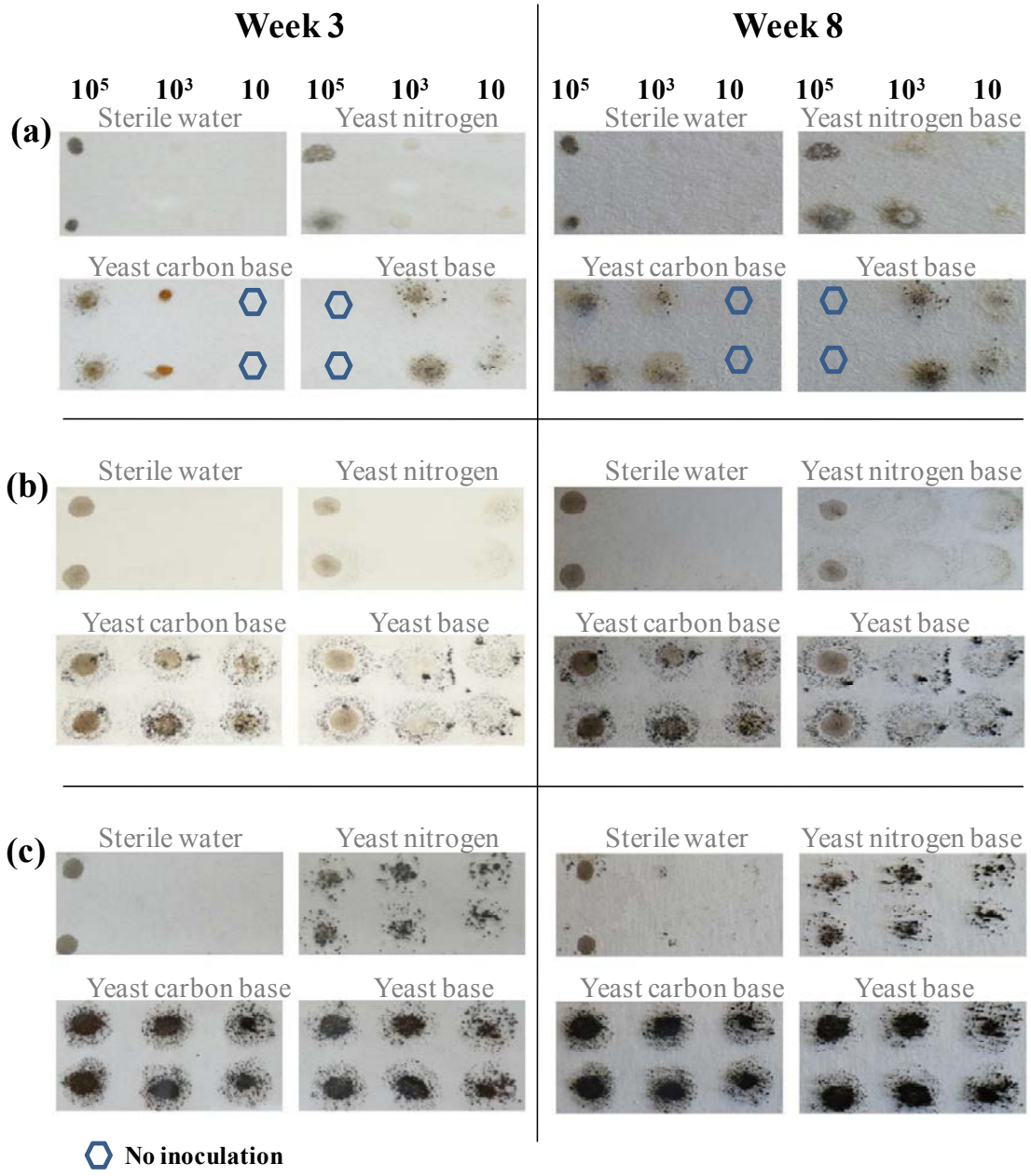


Figure 3. *A. niger* growth on (a) G-Drywall; (b) C-Ceiling; and (c) C-gypsum in 10 X strength nutrient solution on Week 3 and Week 8 of incubation period

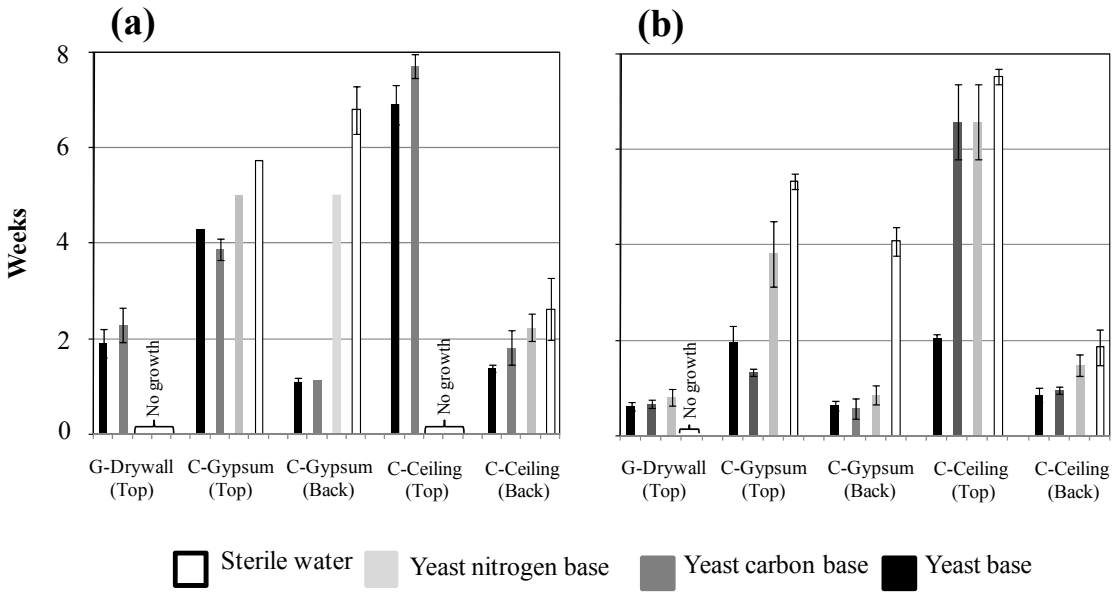


Figure 4. Time until the first observation of mold growth after *A. niger* conidial inoculation of materials in 1X strength nutrient solution for (a) $8x10^1$ spores/inoculated spot, (b) $8x10^4$ spores/inoculated spot. Error bars represent one standard deviation

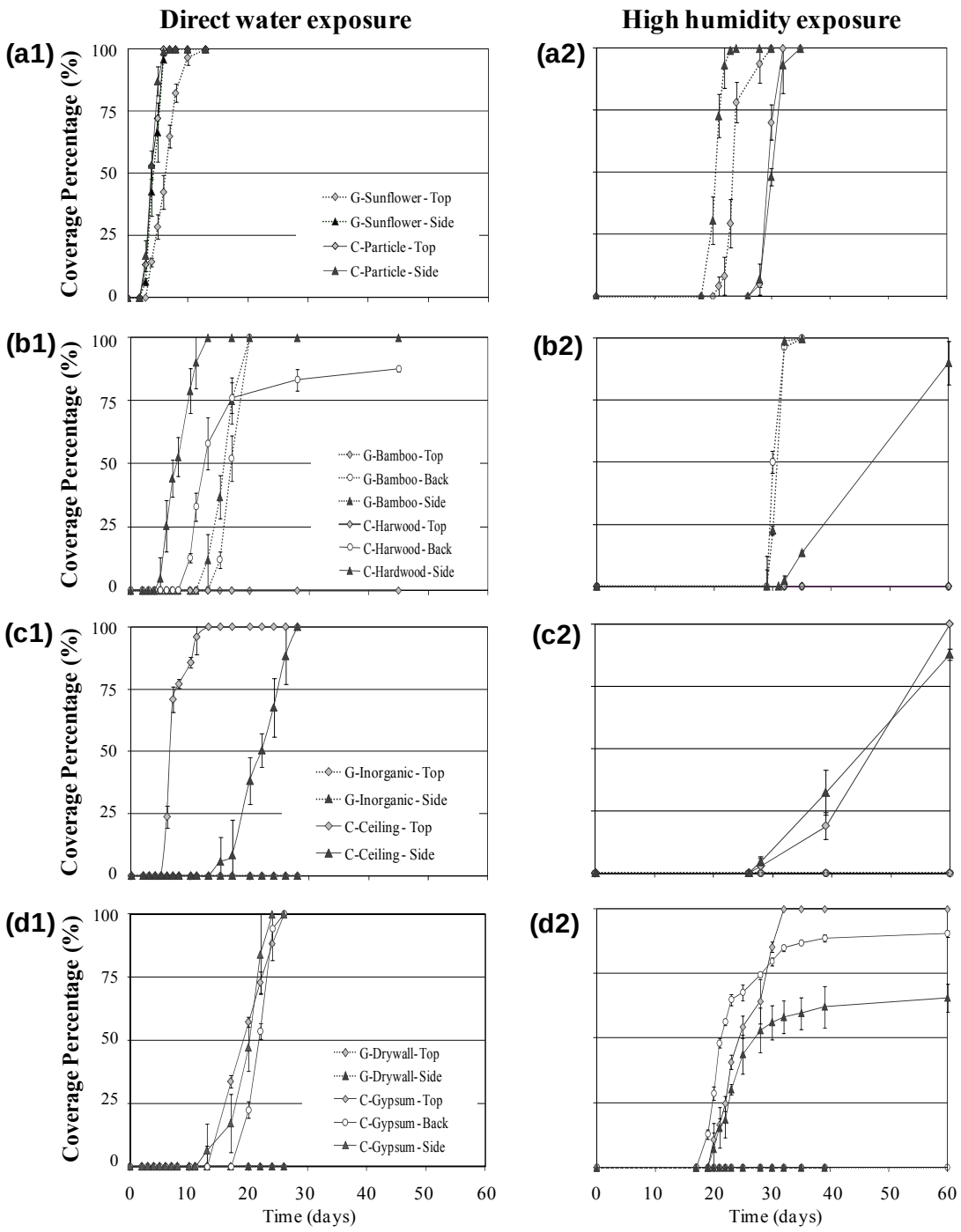


Figure 5. Mold coverage on surface materials of (a) cabinetry (G-Sunflower & C-Particle), (b) flooring (G-Bamboo & C-Hardwood), (c) ceiling (G-Ceiling & C-Ceiling), and (d) wall-board (G-Drywall & C-Gypsum). Error bars represent one standard deviation

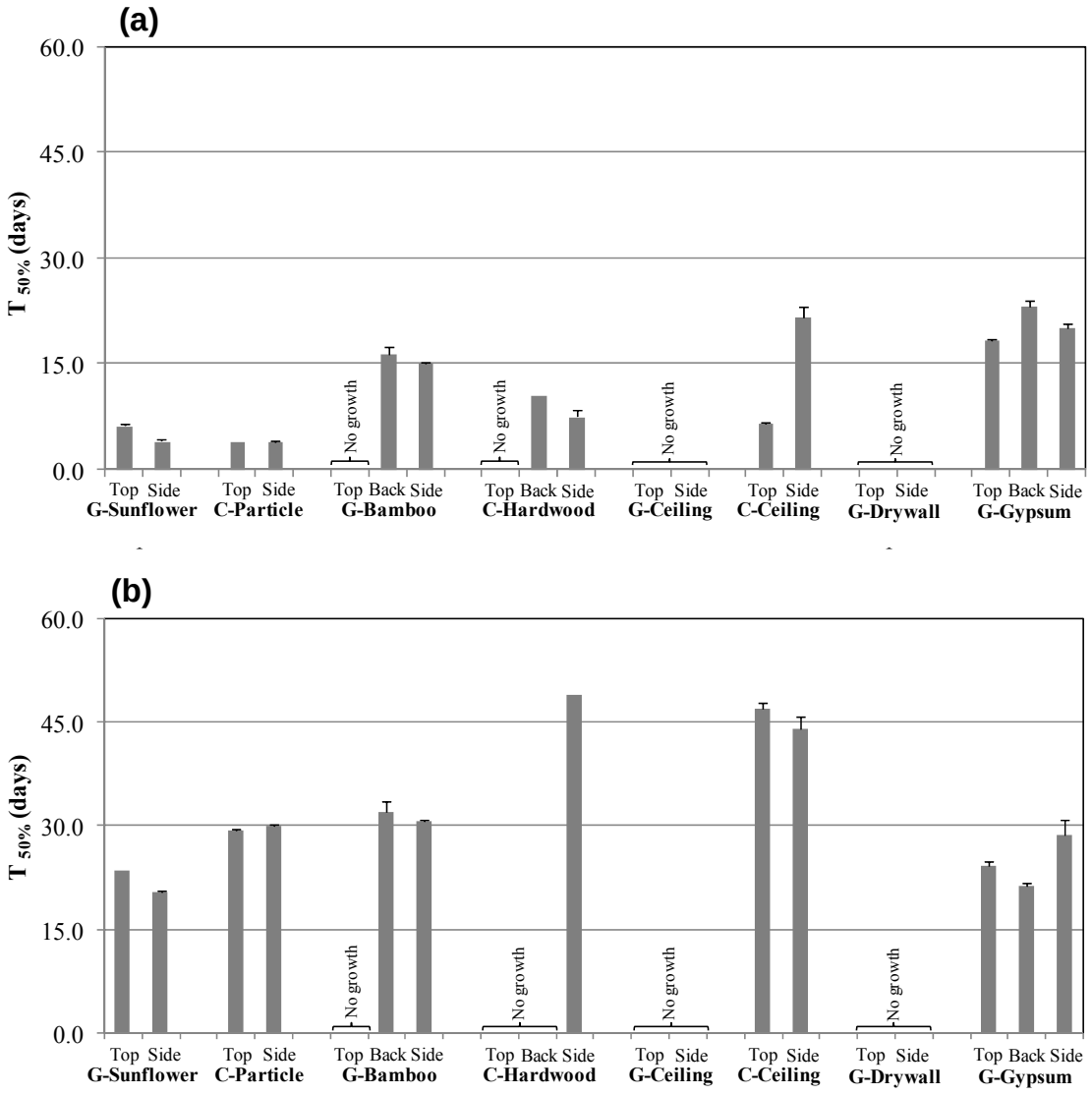


Figure 6. Time until 50% of total material area was covered by mold for (a) direct water exposure or (b) high humidity exposure experiments. Error bars represent one standard deviation

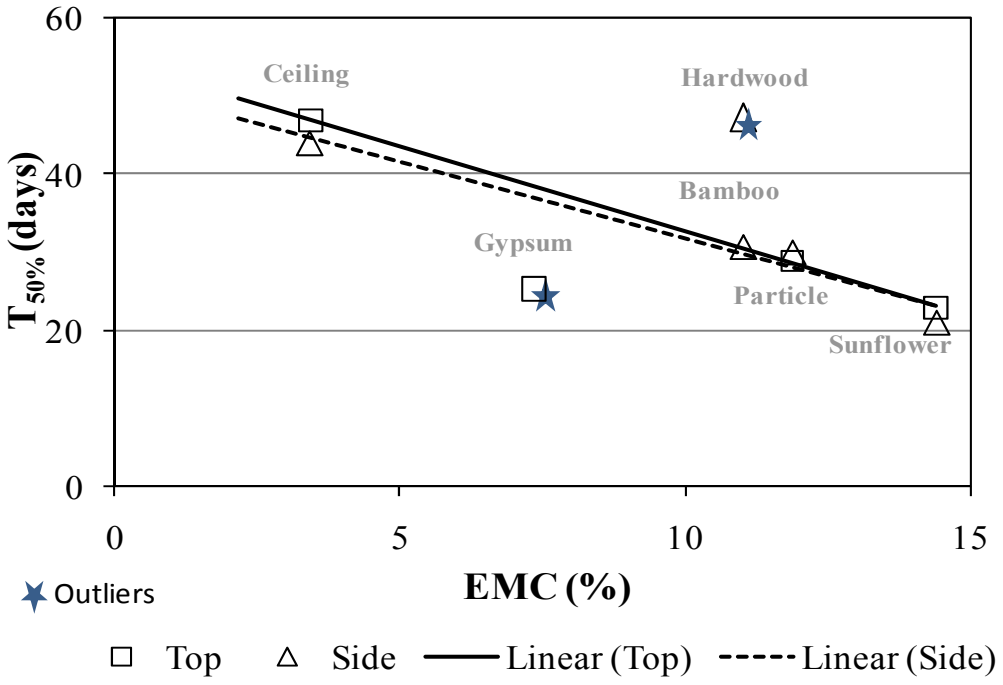


Figure 7. Correlation between EMC at RH of 85-92% and $T_{50\%}$ on the top and side surfaces of test materials

Supporting Document

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Comparison of an Image Processing Method and the ASTM Mold Index for Assessing Mold Growth on Building Materials

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Summary

Six different building materials were naturally inoculated by exposing them to an indoor environment containing a mixture of fungi, After being saturated with water, the wetted materials were incubated for 8 weeks in a closed chamber (RH=90-95% and T=30°C). The mold growth rates were evaluated using the ASTM mold index and an image processing method of our own design. The ASTM index provided a rough assessment of the extent of mold growth on material surfaces and it was found to be less time-consuming than the image processing method. However we found the ASTM rating to be somewhat ambiguous and dependent on an experimenter's judgment. Also, in comparison to the image processing method, the ASTM mold index did not provide a full assessment of the rates of mold growth. Our results suggest that the image processing method may be an attractive alternative to the ASTM mold index for assessing fungal growth on building materials throughout the mold's growth cycle.

Keywords: ASTM; Image processing; Mold growth; Building materials

Introduction

Mold growth on building materials is of increasing concern with respect to protecting occupant health and preventing the deterioration of building materials [Ezeonu

et al. 1994, Klamer et al. 004, Lee et al. 2006]. Many different methods have been utilized to evaluate building materials for their resistance to fungal growth following exposure to moisture. A common assessment method developed by the American Society for Testing and Materials (ASTM) uses visual observation to rate the surface area of a given material that is covered by fungi [Klamer et al. 2004, Vasudeva 2004]. ASTM methods can be used to rate fungal growth on a range of materials including interior coatings (ASTM D3273), synthetic polymeric materials (ASTM G21-2002), paint film (ASTM D3274), and adhesive film (ASTM D4300). In general, specimens are first incubated in a moist environmental chamber in the presence of common indoor fungal species and then examined visually and/or with a microscope to rate the extent of surface fungal growth on a 0-4 or a 0-10 scale. However, the ASTM rating may be considered subjective and possibly too dependent on an experimenter's judgment to decide which index should be applied for each growth rate estimate. Other mold assessment methods which require more effort include using ergosterol content, airborne fungal metabolites or mycotoxins as an indicator of mold growth levels on materials. The objectives of this study were to develop an image processing method for assessing mold growth on building materials and to compare this method with the suggested rating developed by ASTM.

Methods

Three different green materials were selected for this research study: inorganic ceiling tile (G-Ceiling), sunflower board (G-Sunflower), and bamboo flooring (G-Bamboo). A non-green counterpart for each green material was also tested: non-green ceiling tile (C-Ceiling), particle board (C-Particle), and hardwood flooring (C-Hardwood)**.**

When building materials have direct and prolonged contact with floodwater or leaking water, the materials become saturated or nearly saturated and thus they are usually more susceptible to fungal growth. The experimental set-up for this task simulated what happens after a material is submerged in water and then remains in a high humidity environment. Prior to saturation, the six test materials were sterilized and left unprotected in a residential house to allow natural fungal inoculation over a period of 10

days. No additional inoculation with fungal species was made to the material samples. After exposure to ambient air, the material specimens were submerged in sterile water for ten hours. The wetted samples were then incubated for two months in a conditioned chamber at a constant temperature of 30°C and RH of 90-95%.

Fungal growth on the top and side surfaces of each material was visually monitored periodically over a period of two months. Two different assessment methods were utilized: visual observation and rating using the ASTM G21 mold index rating of 0 to 4 as follows: 0: No growth; 1: Traces of growth (less than 10 % of surface area); 2: Light growth (10 to 30% of surface area); 3: Medium growth (30 to 60% of surface area); 4: Heavy growth (60% to complete coverage). An image processing method was developed and implemented as follows: (1) digital images of mold growth were taken using a 7.2Mgp Panasonic Camera (Lumix DMC-LZ6) (2) identification of the fungal contaminated areas on the images; (3) use of the ImageJ software package, developed by the U.S. National Institute of Health (NIH), to determine the area of the whole sample (A_s - m^2) and that of the area contaminated (A_c - m^2); (4) determination of the percentage of area covered by mold growth (P_c - %) which was defined as $P_c = A_c / A_s$. A rating of no or little growth (<10%) was confirmed by microscopic observation (30X).

Discussion and Results

Mold growth levels on the naturally contaminated materials were determined visually and rated using the ASTM scale of 0 to 4. The growth rates of mold on the top and side surfaces of each material are reported separately in Figure 1. Mold growth was observed on several materials during the first week of incubation (Figure 1.a1, 1.b1, 1.c1, 1.d1). Sunflower board appeared most susceptible to mold. For example, by 6 days of incubation, substantial fungal biomass covered almost all the surfaces of the sunflower board. An excessive mass of mold was also observed on the particle board after one week. Silky mycelium grew heavily along the sides and top surfaces of particle board, and black conidia were found later during the incubation period. Of note was the observation of mold growth on only 10% of the sides of the ceiling tile (C-Ceiling) but on the entire top area after the first week of incubation. In contrast, the opposite occurred with the bamboo and hardwood flooring tiles. After 2 months of incubation, almost the

entire area of coated surfaces of both flooring materials was free of mold, whereas all of the side areas were covered heavily by a variety of different colored mold species. This result suggests that a coating layer can be used as a protective layer against mold growth, but that the sides/back of materials may still support such growth. Not surprisingly, no growth was found on inorganic ceiling materials. Even after 2 months of incubation, minimal mold growth was observed on these non-cellulose materials.

Figure 1 also shows the coverage percentage of mold on six green and non-green materials estimated using the image analysis method. The data make it clear that after the spores started to germinate, the mold growth invaded the materials very quickly. For some materials, such as particle board and sunflower board, the entire surface area was covered by mold within one week after mold was first observed. In Figure1.a2, for example, the mold growth rate of sunflower board and particle board rapidly increased from 0% to 100% within the first 5 days of incubation. In contrast, most of the other materials needed more time to be covered by mold.

The two different evaluation methods, the ASTM mold index and the image processing method of our own design, presented both advantages and disadvantages with respect to evaluating the susceptibility of building materials to mold growth. The ASTM mold index approach was less time-consuming and was able to provide a quick and rough assessment of the extent of mold growth on material surfaces. However we found this rating system to be somewhat ambiguous and quite dependent on an experimenter's judgment to decide which index should be applied for each growth condition. Also, the ASTM mold index did not present as full an assessment of the mold growth rates as did the image processing method. Evidence for this is provided by the results depicted in Figure 2, which shows that the ASTM growth index did not reveal the actual growth rate of mold on materials although it generally followed the same trend as the image processing method. For instance, growth coverage higher than 60% cannot be evaluated utilizing the ASTM mold index. In contrast, the slow growth rate of mold observed after 80% of a material's surface was occupied by fungi is clearly evident using the imaging method. This slower growth rate may be due to a limitation in nutrient supply. Our results suggest that the image processing method described here may be an attractive alternative to the ASTM mold index for assessing fungal growth on building materials throughout

the mold's growth cycle. However, image processing proved to be more time consuming and may be less accurate for coverage levels of less than 10%, since the fungal growth and density patterns on some materials are unevenly distributed and difficult to quantify.

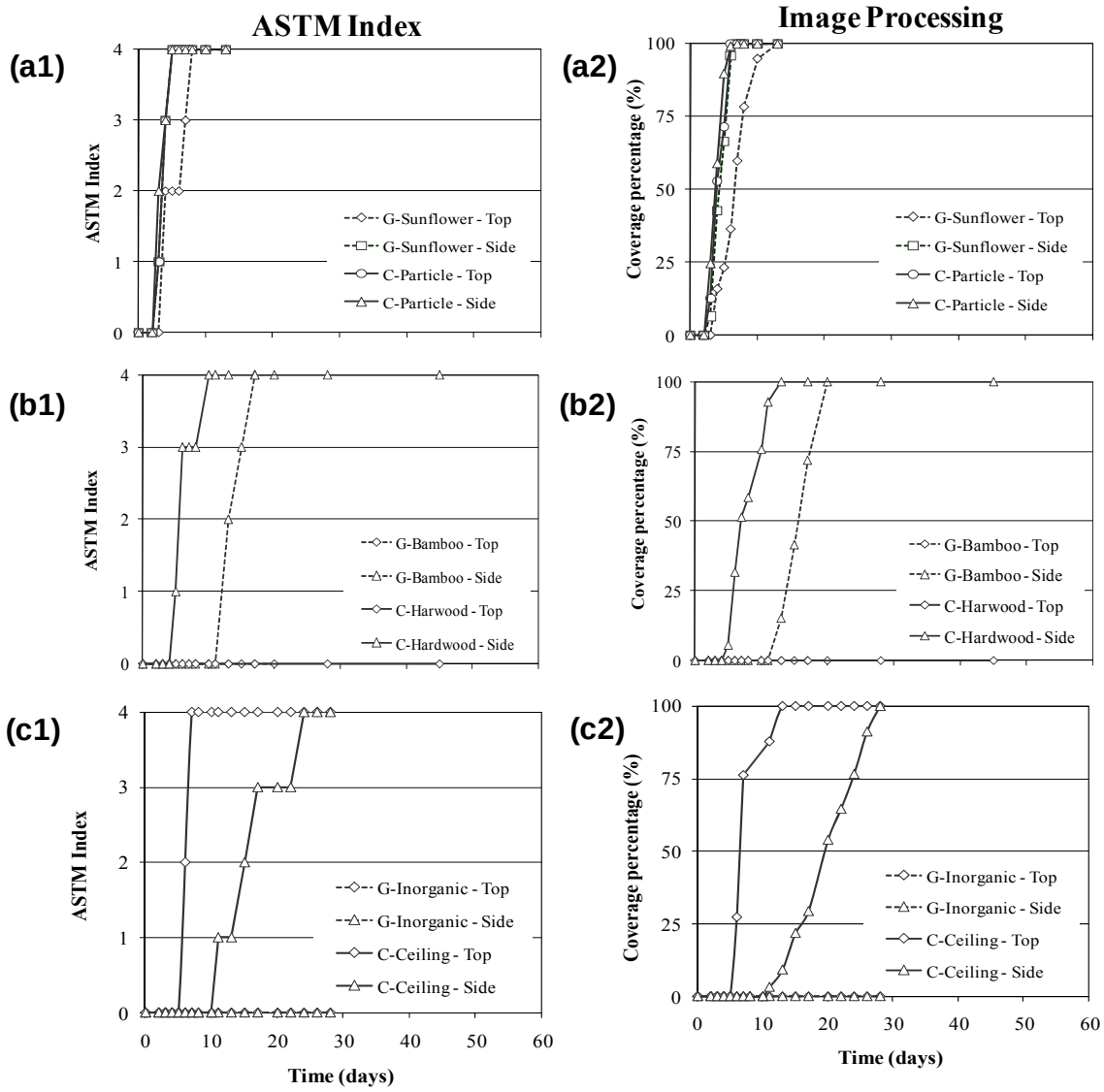

Figure 1. Evaluation of mold coverage using ASTM Index and Image Processing for: (a) G-Sunflower & C-Particle, (b) G-Bamboo & C-Hardwood f, and (c) G-Ceiling & C-Ceiling.

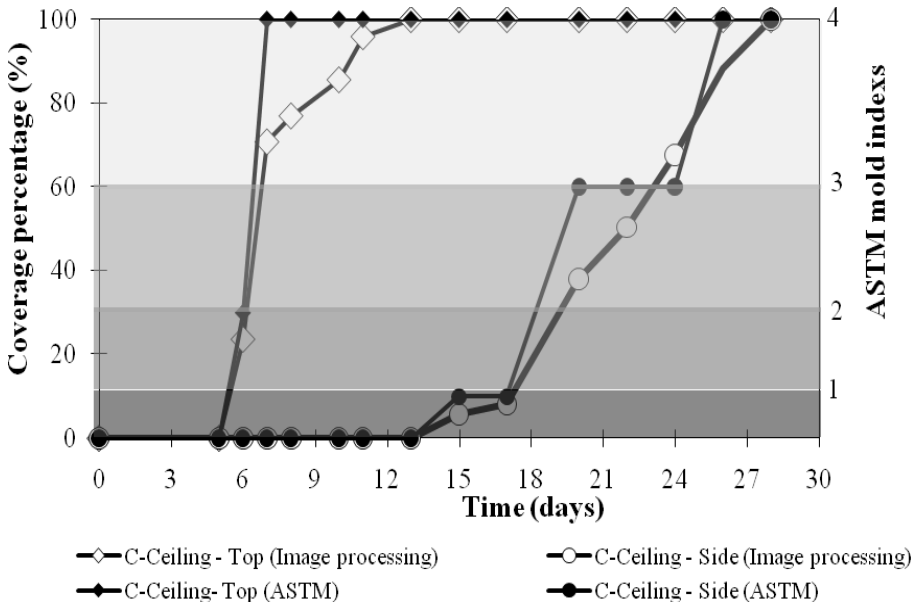

Figure 2. Evaluation of mold coverage on the side and top surfaces of C-Ceiling tiles, using image processing (open symbols) and ASTM mold index rated from 0 to 4 (filled symbols)

Conclusions

Both the ASTM and image processing approach were useful for assessing the susceptibility of building materials to mold growth following moisture exposure. For instance, these rating methods both found that heavy fungal growth developed on cellulose rich materials, while paper-free materials, such as inorganic ceiling tiles, supported little to no growth. As compared to the image processing method, the ASTM index appears to be suitable for providing a quick and rough assessment of mold growth on a given building material. However the ASTM rating (from 0 to 4) does not provide a quantitative measure of mold growth rates which may be a useful parameter for modeling fungal proliferation in indoor environments.

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